ISOTOPES AND RADIATION TECHNOLOGY

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ERRATUM

In the "International Study of Terrestrial Applications of Radioisotopic Power Sources," *Isotopes and Radiation Technology*, Vol. 4, No. 2, under the heading Direct Energy-to-Electricity Conversion, page 172, the low electric power, line 4, should be "[<1 mw(e)]" rather than "[<1 Mw(e)]."

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard, R. E. Greene, F. E. McKinney, Joan E. Carden, and F. J. Miller, *Editors*
A. F. Rupp, *Adviser*

Oak Ridge National Laboratory

In Memoriam

Dr. Paul C. Aebersold, a pioneer in the peaceful uses of atomic energy for medical and other purposes, died May 29. He was chief of the Isotopes Branch, Manhattan Engineer District (MED)—the name given to the atom bomb project—from 1946 to 1947 and director of the Isotopes Division, Oak Ridge Operations, U. S. Atomic Energy Commission (AEC), which assumed the responsibilities of the MED on Jan. 1, 1947. Dr. Aebersold and the Isotopes Development Division were transferred to AEC Headquarters, Washington, D. C., in 1957. He was director of the Division of Isotopes Development (DID), AEC, from 1958 until his retirement in 1965.

His scientific background began as an undergraduate at Stanford University, where he was graduated cum laude with an A.B. degree in physics. Later, as a graduate student at the University of California, he was a member of Professor E. O. Lawrence's group, which developed the cyclotron. He participated in the production and application of the first radioactive materials (sodium and phosphorus) administered to human beings in the late 1930's. Following the receipt of his doctorate in biophysics in 1938, with Phi Beta Kappa honors, Dr. Aebersold continued his work with radioisotopes and also investigated the properties of biological reactions resulting from fast-neutron beams. As a research associate in the Radiation Laboratory at Berkeley from 1940 to 1942, he was in charge of operating the 60-in. cyclotron.

In January 1946, Dr. Aebersold was asked to transfer from Los Alamos Scientific Laboratory to Oak Ridge, to become chief of the Isotopes Branch, MED. His earlier experience with the cyclotron group at Berkeley made him a logical choice to direct this first peaceful distribution and use of atomic energy.

During the first 7 months of 1946, the new Isotopes Branch was a beehive of activity. Dr. Aebersold wrote memorandums for his superiors in Oak Ridge, flew to Washington to be on hand when the memorandums were delivered, and prepared replies for the return classified mail to Oak Ridge. This facilitated a smooth and rapid beginning for public release of these "secret" wartime materials.

Key members were added to the staff, administrative policies and procedures hammered out, application and authorization forms for radioisotope procurement designed, and review committees established. The first catalog, "Availability of Radioactive Isotopes—Announcements from Headquarters, Manhattan Project, Washington, D. C.," was published in *Science*, 103: 697-705 (June 14, 1946).

All of these efforts and policies were highlighted by the first shipment of radioisotopes on the afternoon of Aug. 2, 1946, from the Oak Ridge Graphite Reactor. This first peacetime product of the government's huge atomic energy facilities was a pea-sized unit of ^{14}C (as $BaCO_3$) which weighed about 3 mg and contained 1 mc of radioactivity. The first shipment went to the Barnard Free Skin and Cancer Hospital, St. Louis, Mo., for research and study of the processes by which cancer is produced.

Since the first shipment in 1946, isotopes have found wide application in the fields of biology, medicine, industry, agriculture, the physical sciences, engineering, etc. In 1966, more than 2.5 million curies of processed radioisotopes were distributed by Oak Ridge National Laboratory (ORNL)—one of approximately 83 foreign and domestic suppliers today. Much of the credit for the worldwide growth in the everyday applications of isotopes has resulted from the imagination and insight that Dr. Aebersold gave to the program.

Section

I

Isotopes and Radiation Technology

Isotope Technology Development

Mail-Sorting Apparatus Using Radioisotope-Generated X-Ray Fluorescence*

Abstract: A mail-sorting system based on X-ray fluorescence generated by the bremsstrahlung from a ^{147}Pm/Al source was developed. The prototype unit was operated by a technician with limited training and handled 10^7 letters in a 500-hr period without difficulty. Production units could be built for about $5000.

The first step in automated mail sorting is turning all the envelopes in the same direction so that the addresses can be read. If the stamp is in the upper right-hand corner, which is an almost universal practice, the sorting can be controlled by devices that are sensitive to materials in or on the stamp. High-rate systems based on the use of ultraviolet-fluorescent materials in the stamp are in operation, and other techniques have been suggested, e.g., addition of ultraviolet-phosphorescent materials to the stamp, use of magnetic ink, and insertion of an optical contrast medium between the stamp and envelope. However, all these techniques have disadvantages — the ultraviolet-sensitive additives color the stamp paper, magnetic ink is very dark colored, and the optical contrast technique is unreliable.

A study was therefore begun in 1964, by L'Atome Industriel under a contract with the French Centre d'Étude des Télécommunications, on a system based on radioisotopes. An apparatus was developed, and a prototype unit was constructed and tested. The prototype unit, which is described below, met the specifications set up by the Post Office Department:

Sorting speed	36 thousand letters/hr
Area to be examined	50 by 40 mm
Errors when stamp is absent	1 in 10,000
Errors when stamp is present	1 in 100

*Details are given in "Application of X-Ray Fluorescence to the Automation of Mail Sorting," in French, by G. Robin and E. Darigny, pp. 152–172, Vol. 1, *Radioisotope Instruments in Industry and Geophysics*, Symposium Proceedings, Warsaw, 1965, International Atomic Energy Agency, Vienna, 1966; and in English by E. Darigny and G. Robin, pp. 786–804, Proceedings of the 2nd Symposium on Low-Energy X- and Gamma Sources and Applications, University of Texas, Austin, 1967, Report ORNL-IIC-10, P. S. Baker and M. Gerrard (Eds.).

In a 500-hr test, the unit handled 10^7 letters. Errors were within specified limits, and the envelopes were turned correctly. A Post Office employee with no previous experience in nuclear electronics operated the equipment after a short training period, and there were no operating difficulties.

Prototype-System Description

In the mail-sorting system developed by the French firm, fluorescence X radiation is produced by impingement of the bremsstrahlung from a ^{147}Pm/Al source on zinc oxide added to the paper pulp from which the stamp is made. The ^{147}Pm/Al source was chosen because it has a convenient half-life, is low in cost, and is readily available. The noise-to-signal ratio of the system will permit operation at a rate of 60 to 70 thousand letters per hour if the mechanical parts of the equipment will hold up at this speed. Zinc oxide was chosen because the zinc 8.6-kev fluorescence X ray can be detected conveniently. It is inexpensive, does not change the color or strength of paper to which it is added, and in the amount required (1 mg/cm^2) would not introduce any toxicity problems. The source activity and location with respect to other parts (Fig. 1) were selected to give maximum response

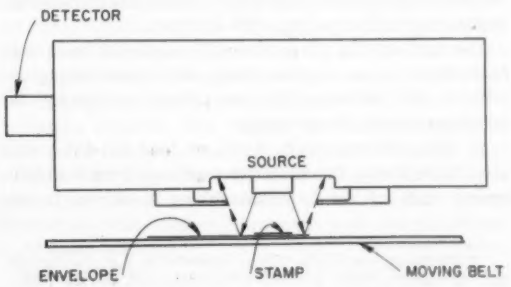

Fig. 1 Relative positions of source, counter, and stamp.

to the fluorescence X radiation when a stamp passes in front of the detector. The detector is a proportional counter filled with argon–methane. Its beryllium window is rectangular, with dimensions slightly greater than those of the envelope area examined.

The prototype mail sorter is made up of four units because, assuming that the stamp is placed in the upper right-hand corner, there are four possible positions of the envelope in the sorter (Fig. 2).* Each unit contains a sensing head with two 2-curie $^{147}Pm/Al$ sources in a backscattering geometry, a detector, a recorder, and the usual electronic circuits.

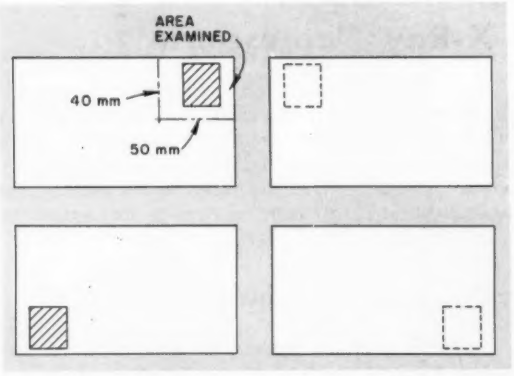

Fig. 2 Possible positions of envelope in sensing head if stamp is in upper right-hand corner.

The electronic circuits are standard French modular transistorized units. One unit (Fig. 3) includes a high-voltage supply for the detector, a preamplifier in the detecting head, a single-channel analyzer, a gating circuit, and a preset scaler. When a letter enters the sensing head, the gate opens under the control of a photocell, and each unit examines its assigned area. The scaler records energy pulses of the 8.6-kev K X radiation from the zinc, about 15 msec being required. If the preset count is obtained, an output pulse is generated. The unit is reset by entry of the next letter. If no stamp is present, the scaler records background pulses due principally to the backscattering of X rays from the envelope.

The test system (Figs. 4 and 5) contained two pulse generators—one to give gating pulses simulating the letters and the other to give pulses simulating the count generated by the stamp.

In the prototype unit, 4 cm of lead shielding was used to decrease the radiation exposure from the high-energy tail of the bremsstrahlung spectrum to the tolerance level of 0.25 mr/hr at the surface of the sensing head. A lining of 1-mm aluminum was placed inside the cover of the unit to reduce the background from backscatter of the source radiation on lead.

(Martha Gerrard)

*If the stamp should be placed elsewhere, e.g., in the upper left-hand corner, it would not be detected, and the letter would be routed for manual sorting. This would, of course, apply to any sorting system based on stamp examination.

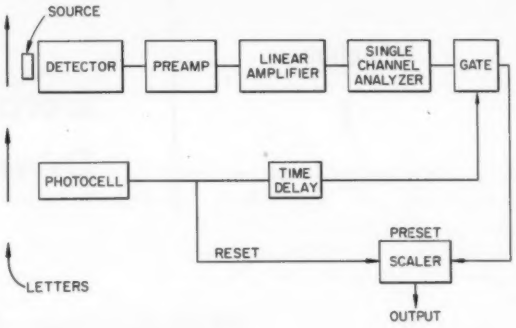

Fig. 3 One unit of the mail-sorting system.

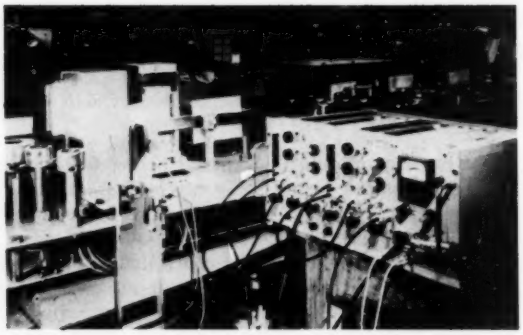

Fig. 4 Detecting heads and electronic circuits of X-ray system.

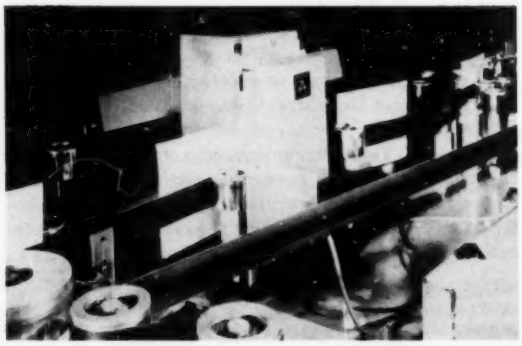

Fig. 5 Closeup view of detecting head.

A Review of the Use of Radioisotope Tracers for Evaluating Parameters Pertaining to the Flow of Material in Plant and Natural Systems*

By K. Ljunggrent

Abstract: The merits of radioactive isotopes as compared to other substances (salts, dyes) for use as tracers have been discussed at length by many authors and are now widely appreciated. For industrial plant investigations they are particularly useful because of (1) the great sensitivity of detection which permits the use of such small amounts of tracer material that there is no possible interference with the process studied and (2) the availability of gamma-emitting nuclides of many elements that in most cases make possible direct measurements of tracer concentrations from the outside of vessels and pipes carrying the tracers.

The following uses of radioactive tracers in studies of flow of material, especially of fluids, are typical:

1. In experiments designed to provide data for fundamental flow-theory formulation

2. For measuring flow parameters under well-defined conditions of flow: velocity, volumetric flow rate, dispersion in flow, etc.

3. For general examination of the characteristics of a flow system with respect to flow patterns, flow paths, etc.

4. For measuring the dilution caused by the dispersion of dissolved or suspended matter

5. For the evaluation of process and process-equipment performance by arranging tracer tests, which provide residence-time distributions (from which mean residence times, dispersion indexes, blocked volume, hold-back, and similar characteristics can be extracted), information on mixing properties, channeling, recirculation, unintended connections, leakage, etc.

The principles of the various methods and the practical procedures are discussed; when applicable, comparisons with alternative techniques are made, and reported figures on attainable precision and accuracy are cited. The theoretical aspects of the significance and evaluation of tracer investigations of material turnover are dealt with, with particular reference to concepts and models used in analyzing residence-time distributions. An attempt is made to discern general trends of development in radiotracer applications, such as the use of the obtained information for process control purposes.

Radioactive substances have added considerably to the potentiality of the tracer method since they provide suitable tracers for almost any purpose. The radioactive tracers can generally be detected at very low concentrations, and, if they emit gamma radiation, they can be measured directly on-site through pipe walls, etc., or in large volumes of matter. "Chemical" tracing, which involves tracing of an element, is often limited by a narrow choice of radioactive nuclides with suitable radiation characteristics, whereas "physical" tracing, where objects or phases are subjected to tracing, usually permits the selection of a tracer nuclide with radiation characteristics tailored to meet the specific requirements of an on-site continuous measurement of tracer concentration.[1]

In the use of on-site measurement methods, all interferences with the process being studied are avoided, except for those which may occur at the point of tracer administration or at structures introduced for internal detectors. However, with care the tracer can generally be introduced in such a way as to make these disturbances negligible. Moreover, the radiation measurement is specific in nature and very little affected by environmental conditions.

Studies of material transport and determination of parameters characteristic of the flow of fluids and solids have been made possible or facilitated by the use of radioactive substances as tracers. Numerous studies of this kind in industry and in natural systems have been reported in the literature.[1–5]

Studies of Fundamental Flow Characteristics

The properties of fluid flow have been treated theoretically by a number of authors. Extensive analyses on dispersion of a soluble matter in a stream of solvent have been published by Taylor[6–8] and modified or extended by others, inciuding Aris[9–11] and Tichacek, Barkelew, and Baron.[12] These theories have been checked experimentally by using tracers of various kinds—often radioactive materials. Turner[13] reviews the theory and some experiments, and in this connection an early investigation by Hull and Kent[14] can be mentioned in which ^{140}Ba and ^{124}Sb were used as oil-soluble compounds for studying oil flow in pipelines. An investigation by Lee[15] on a partially "silted-up" pipe may also be mentioned as a good demonstration of the practical usefulness of information calculated by elementary means from simple tests.

Beatty, Ferrell, and Richardson[16] have concerned themselves with studies of velocity distributions over a flow cross section with a view to establishing in particular the characteristics of the boundary layer. Sodium-24 was used as a tracer in a series of laboratory investigations.

Clayton, Webb, and Whittaker[17] have used ^{85}Kr to study the longitudinal dispersion of a gas in turbulent flow along a pipe. Results agreed with those predicted

*The material in this article was presented as paper SM-84/22 at the symposium in Prague, Nov. 21–25, 1966, on *Radioisotope Tracers in Industry and Geophysics*, sponsored by the International Atomic Energy Agency and is published by the kind permission of the Agency and the author.

†Isotope Techniques Laboratory, Stockholm 0, Sweden.

by a theoretical modification of Taylor's theory, if the compressibility of the medium is considered.

Tracer experiments have also been conducted in open-water courses, rivers, and canals in efforts to establish more or less generally valid dispersion data. Hull[18] reported from measurements on the American River near Sacramento, Calif., that the longitudinal dispersion coefficient, D, could be related to the volumetric flow rate, v, and the linear velocity, u:

$$D = 2.5\ (vu)^{1/2} \tag{1}$$

Clayton and Smith[19] have studied the dispersion in rivers with flow rates ranging from 2.5 liters/sec to 2 m^3/sec, and Timblin and Peterka[20] give some information on longitudinal dispersion in a canal conveying some 10 m^3/sec. In both cases the corresponding mixing length is far greater than that which can be derived from Hull's formula.

Kato et al.[21] report on several determinations of dispersion coefficients in the Sorachi River in Japan.

Flow-Rate Measurements

Radioactive tracers are used when the flow rate of liquids and gases must be determined precisely, when occasional data concerning flow rate are needed, or when calibration of installed conventional flowmeters is required.

There are four established standard methods; these are[22]

(a) Velocity (peak timing) method
(b) Total-count method
(c) Continuous (total) -sample method
(d) Constant-rate-injection (dilution) method

The first three are based on a sudden addition of tracer to the stream, while the fourth, as indicated by its name, involves a continuous and constant rate of injection.

The velocity measurement method (a) is begun by injecting a small volume of radioactive fluid into the stream in a closed conduit. On passing two detectors some known distance apart, the slug of tracer gives rise to two response pulses on the associated count-rate meters, differing in time by an amount T. If the internal cross section of the conduit S (or more strictly the volume, V, of the fluid between the observation points) and the distance, L, between the detectors are known, the volumetric flow rate, v, can be calculated:

$$v = L \cdot S/T = V/T \tag{2}$$

For a gaseous system, pressure and temperature conditions must also be known.

The total-count method (b), which was developed by Hull,[23] is based on a balance calculation on a momentarily added amount of tracer; the calculation is made possible by the observance of the passage of the tracer at a downstream point with a detector connected to a scaler.

The tracer balance yields (cf. Eq. 47)

$$Q = v \int_0^\infty c(t)\ dt \tag{3}$$

where Q is the amount of tracer added to the stream, v is the volumetric flow rate, and $c(t)$ is the concentration of tracer at the point of observation (which varies with time, t). Since the rate, r, at which counts are accumulated on the scaler at any given time, t, is proportional to the concentration of the tracer, then

$$Q = \frac{v}{F} \int_0^\infty r(t)\ dt \tag{4}$$

where F is the proportionality constant. The integral is the total number of counts, N, above background, registered by the scaler during the passage of the tracer. Therefore

$$Q = v \cdot N/F \tag{5}$$

or*

$$v = Q \cdot F/N \tag{6}$$

No determination of absolute activity is required since a calibration sample diluted from a known volume of the injection solution can be used to determine F by an additional measurement.

The continuous-sample method[24] (c) is a modification of the total-count method. Samples are drawn continuously or intermittently from a point downstream from the injection, instead of being measured in situ as in the total-count method. If a continuous sample is taken over a time period, T, and, after careful homogenization of the sample, its activity is measured in terms of the counting rate, $\bar{r}$, in a counter setup having an efficiency F, we obtain

$$v = QF/\bar{r}T \tag{7}$$

The analogy with Eq. 6 is evident, the integral $\int_0^\infty r(t)\ dt$ being evaluated as (mean counting rate) × (period of sampling). Of course, T must be long enough to encompass the whole duration of the tracer transport past the sampling site.

In the constant-rate-injection method (d), tracer solution is added at a constant and measured rate to the stream being gaged. After steady tracer concentration has been reached at a downstream point sufficiently distant from the injection to provide for lateral mixing, one or several samples are withdrawn from

*In Hull's original presentation the amount of tracer was denoted by A and the volumetric flow rate by Q, which gives the better known formula $Q = AF/N$.

the stream and measured for tracer content. Conservation of tracer requires that

$$v = v_0 \cdot c_0/c \qquad (v \gg v_0) \tag{8}$$

where v is the unknown flow rate, v_0 is the injection rate of tracer solution having a concentration c_0, and c is the concentration of tracer at the sampling point. If a diluted aliquot of the injection solution is used for determining c_0 in the same counting setup that is used for assessing c, no absolute tracer concentrations need be determined, and c_0/c can be replaced with r_0/r, the counting rate ratio for samples taken from the injection solution and from the stream, respectively.

The velocity method is best suited for measurements of turbulent flow in closed conduits, while the other methods are equally applicable to flow in open channels and streams. The velocity method also requires that the flow fill the entire cross section of the conduit.

A requirement common to all the methods is that the distance between the point of injection and the point of measurement (with the peak-timing method, the first observation point) must be large enough for lateral mixing to have occurred. While this is an indispensable condition for in situ measurements where a considerable fraction of the cross section of the flow may contribute simultaneously to the detector signal, Evans and Ely[25] have shown that, when measurements are confined to one single point (as with sampling), a sufficient condition is that the time integral of tracer concentration be constant on an arbitrary surface that includes the measuring point. The time integral will, of course, be constant if the concentration itself is constant on such a surface.

The question of mixing length has been discussed by Clayton,[22] who has also suggested various ways of decreasing it by using multiple-point injection and by including circulating pumps in the test section.[26] For measurements of pipe flow, a mixing length of 300 diameters is considered adequate, and a length of 100 diameters is usually sufficient to limit to ~1% the contribution to the error in the final flow-rate value from this source. Use of mixing pumps can reduce the mixing length to 20 to 30 pipe diameters.

Under favorable conditions an accuracy of 0.1 to 0.5% is attainable in practical operation with both the velocity and the constant-rate-injection methods for measuring liquid flow; this is as good as can be achieved by weighing the discharge.[27] A normal accuracy in gas flow-rate measurements using the velocity method[27] would be about 1%.

Without much effort all the described methods except that of total count can be operated to a precision of 1 to 2%; with the total-count method, the precision attainable is a few percent. The high precision makes these methods eminently useful in a large number of industrial operations as well as for hydrological purposes. The relative merits of the methods for various practical tasks have been discussed by Clayton[22] and by Mott.[28] Because the velocity method is easy to operate—particularly if repeated measurements are to be made—and requires no calibration, one can say that this method is more convenient than other methods for measuring flow rates in closed conduits if an adequate length of straight pipe without disturbing valves, orifice plates, bends, etc., is available.

For open flow systems, or for closed systems where the velocity method cannot be conveniently applied, one of the other methods is used. The velocity method and the constant-rate-injection method can be used together in closed conduits where the internal dimensions are not accurately known. A few simultaneous runs calibrate the velocity method. The calibration is then used for repeated measurements to save time and labor.[22]

In addition to the velocity method, the total-count and the total-sample methods[28] are of importance in petroleum-refinery operations. The constant-rate-injection method is in wide use for measuring industrial effluent[29] flow rates of 0.1 to 5 m^3/sec and cooling-water rates of 5 to 10 m^3/sec in thermoelectric power stations.[30] The use of the latter method can be extended to greater flow rates, but with increasing flow, the total-count, and, in particular, the total-sample methods become more and more attractive since they are more economic in respect to tracer consumption for a given level of precision. Timblin and Peterka[20] report on the use of the total-count method for measuring canal flow rates in excess of 200 m^3/sec. In India, attempts have been made to measure river flow rates[31] up to 5000 m^3/sec.

At an early stage the velocity method was used to measure pipeline flow. A recent in-plant application has been the measurement of acid circulation in a hydrocarbon alkylation plant with ^{198}Au (in sodium cyanide solution) as a tracer.[32] Flow-rate measurements on riser and downcomer tubes in a large boiler have been determined by the velocity method with the use of $NH_4{}^{82}Br$ (Ref. 33). Gas flow in a synthetic ammonia plant has been measured with ^{85}Kr by using the total-count principle,[34] and steam flow in a refinery superheated-steam line by using continuous sampling with ^3H_2O (Ref. 35). A variation of the total-count method is the carrier-gas method, which can be used as an alternative to the total-sample method; it employs a noncondensable but, in other respects, convenient tracer gas like ^{85}Kr to measure flow of condensable gases—e.g., steam.[36] Recently an account has been given of measurements of leakage rates across an oil trap in a carbon dioxide knockout pot using the total-sample method and $^{14}CO_2$ as a tracer.[37] A detailed discussion on the use of radioactive tracer methods for measuring gas flow rates has been presented by Johnson.[38]

Other work on flow-rate measurements in canals and natural streams has been reported by Clayton and Smith,[19] Hull,[39] Guizerix et al.,[40] and Gardner and Dunn.[41]

An IAEA working group on isotopes in hydrology has published a report from a meeting held at Grenoble in October 1965, which gives some detailed information on recent achievements in flow gaging and a number of related subjects.[42] One line of development which is being pursued at the Research Center for Radiohydrometry in Munich[43] is the enrichment of ^{131}I tracer from collected samples to increase the limit of detection of the method and thus to reduce the amount of tracer necessary to measure large flow rates (>100 m^3/sec).

The described methods, at least in principle, can also be used for regularly repeated measurements in a more or less automatic arrangement. Keys, Alexander, and Eichholz[44] have developed a device for measuring the flow rate of fuel to a jet engine in which a quantity of ^{131}I is injected from a supply into the fuel line, and the transit time of the ^{131}I to a detector down the line is recorded. The injector is actuated by a manual switch that also starts the timing circuit.

Flow rates of solids are sometimes of concern in industrial processes. One important application of radioactive tracers for measuring flow rates of solids is the monitoring of catalyst circulation rates in refinery control. The technique, which was one of the first applications of radioactive tracers in a refinery, involves the timing of the passage of a radioactive catalyst bead between counters placed at two or more points in the circulation system.[45]

Studies of Continuous-Flow Systems

THEORY AND EVALUATION METHODS

With the general introduction of continuous instead of batch processes in recent years, the conditions under which solids and fluids are transported through processing vessels have received ever increasing attention. These conditions can be studied by observing the response of the system to spontaneous or deliberately created disturbances at the inlet—for example, to a variation in concentration of one of the components.

An extremely convenient way of effecting a disturbance is to introduce a tracer substance into the feed stream. In this way an artificial disturbance is created which is capable of yielding information on the response of the system to a real change in concentration of process components, without in any way upsetting the production process. The tracer must be amenable to selective detection inside the system or in the effluent stream. If chemical identity is required between the tracer and its "mother substance,"* only isotopic tracers are usable; in other cases any readily detectable substance can be employed if it is physically and chemically compatible with the flowing medium under the process conditions. As usual, radioactive isotopes offer a number of advantages also as physical tracers,[1] the most important being that measurements can be performed on-site, e.g., directly on the pipe from the vessel with no need for sampling.

These tests can be arranged in various ways. The tracer may be added all at once, giving a pulse input that sometimes can be considered to approach a theoretical delta function, i.e., a pulse of infinitely short duration and infinite amplitude such that its area in a concentration-vs.-time diagram is finite and proportional to the amount of tracer added. This is the method which in the majority of practical cases is easiest to engineer and which consumes the smallest amount of tracer for a given quality of result. It is also attractive because the concentration-vs.-time curve recorded at the outlet will yield directly the transfer function of the system (restricting ourselves to linear systems); in other words, the delta function represents disturbances to the system of all frequencies with equal amplitudes.

The tracer may be administered alternatively to give a step input: the concentration of tracer in the inlet is suddenly increased from zero to a certain value and the injection thereafter continued at a constant rate until a steady state has developed. The concentration-vs.-time curve thus obtained at the outlet will be the time integral of the curve that is obtained with a delta pulse input. Experimental conditions, particularly for studying fast systems, may make it simpler to work with a negative step signal that involves rapidly cutting off a steady supply of tracer. Still other methods are the modulation of the injection rate (e.g., sinusoidally) or the use of a random input function that is monitored by a detector at the entry of the system. A demonstration of the techniques is given in Fig. 1, where the response of a system to various input stimuli is shown.

If the system being studied is linear, all these experiments will contain essentially the same information. The possibility of extracting this fundamental information from the experimental results may depend, however, on the experimental procedure adopted. In practice there has been an establishment of certain methods that are preferred for particular tasks since these methods are easier to operate or give higher inherent precision or accuracy for a given consumption of tracer.

The evaluation of residence-time distributions has been of considerable interest since around 1950. An early treatise on residence times in series-connected stirred vessels was given by MacMullin and Weber[47] in 1935. In 1949 and 1952 Gilliland and Mason[48,49] published two papers in which a first outline was given for a method of presenting residence-time curves in

*The term *mother substance* has been adopted from Bergner.[46]

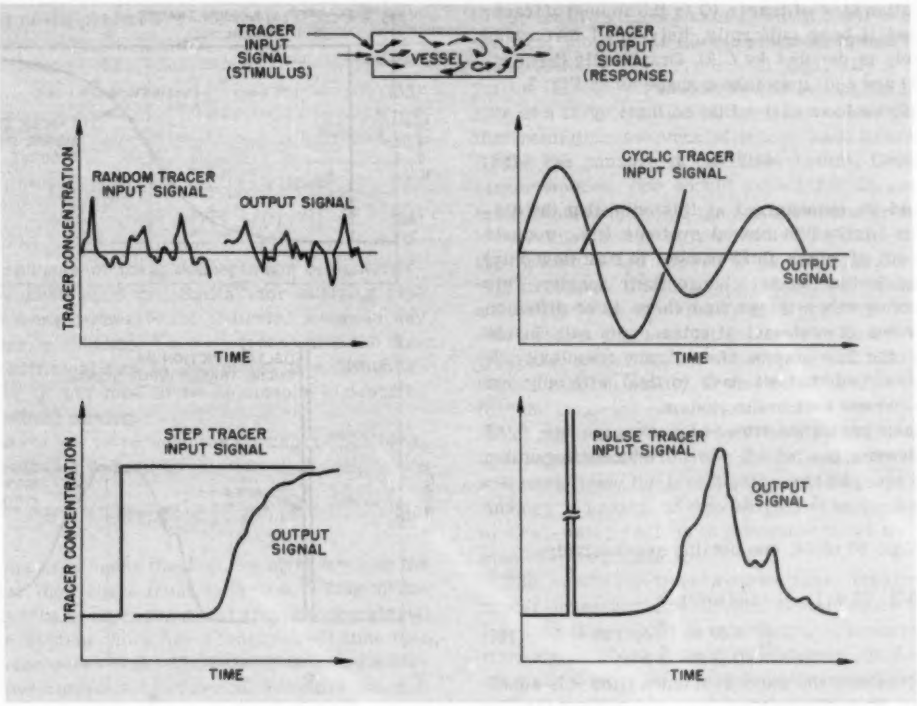

Fig. 1 Response of a continuous-flow system to various tracer input signals. (After Levenspiel, Ref. 51.)

nondimensional quantities; the method was later developed by Danckwerts to an elegant and self-consistent set of concepts in a paper[50] that has become a minor classic in the field. The basic idea of Danckwerts is to introduce age functions that describe the distribution of ages of the material within the vessel and in the effluent stream. The volume, V, of the vessel occupied by the fluid is considered to be constant as is the volumetric rate of inflow and outflow of fluid, v. If V is divided by v, the nominal mean residence time $\bar{t}_n = V/v$ is obtained.* It is very useful to express time in units of the nominal mean residence time $\bar{t}_n$, as has been done by Levenspiel in his modification of Danckwert's concepts[51]

$$\theta = t/\bar{t}_n \tag{9}$$

The internal age distribution, $I(\theta)$, defines the fraction of fluid particles in the system at any instant having ages between θ and $\theta + d\theta$, which is $I(\theta)\, d\theta$. With this definition, $I(\theta)$ is analogous to the frequency function of a stochastic variable.

The exit age distribution, $E(\theta)$, defines the fraction of material with ages between θ and $\theta + d\theta$, at the moment of leaving the system, as $E(\theta)\, d\theta$.

Between $I(\theta)$ and $E(\theta)$ a valid relation is

$$E(\theta) = -dI(\theta)/d\theta \tag{10}$$

Now, $F(\theta)$ is introduced as the function that describes the variation of tracer concentration in the system exit after the introduction of a positive step signal at time $\theta = 0$ in terms of the constant tracer concentration applied to the inlet. With this definition

$$F(\theta) = 1 - I(\theta) \tag{11}$$

$$F(\theta) = \int_0^\theta E(\theta')\, d\theta' \tag{12}$$

$$E(\theta) = dF(\theta)/d\theta = -dI(\theta)/d\theta \tag{13}$$

If, instead, a delta-function tracer input signal is applied to the system and the tracer concentration is recorded at the output as a function of time, a concentration-vs.-time curve is obtained which may be denoted $c(t)$, or in reduced time units, $c(\theta)$. The function $c(\theta)$, normalized with respect to the initial

*A certain confusion is sometimes caused by the use of the term *residence time* for the age distribution in the exit stream, which, according to some authors, should be used only for the age distribution of material inside the vessel; for the exit stream, terms like *transit time* or *time of sojourn* are used.

concentration Q/V of tracer (Q is the amount of tracer added) had it been uniformly distributed throughout the vessel, is denoted by $C(\theta)$. Graphically the functions $F(\theta)$ and $C(\theta)$ may take a shape as in Fig. 2.

For $C(\theta)$ we have $C(\theta) = E(\theta)$ so that

$$\int_0^\infty C(\theta)\, d\theta = 1 \qquad (14)$$

It should be emphasized at this point that the discussion is limited to closed systems (i.e., vessels into and out of which fluid passes in bulk flow only) and to fluids that do not change their density. This means among other things that there is no diffusion or dispersion of material at entrance or exit. Inside the vessel the flow may be of arbitrary complexity. It is also assumed that we have to deal with only one entrance and one exit in the system.

The mean residence time — the average age, $\bar{\theta}$, of the fluid leaving the vessel — is in reduced time units:

$$\bar{\theta} = \int_0^\infty \theta E(\theta)\, d\theta \qquad (15)$$

Using Eqs. 10 to 14, one obtains successively

$$\bar{\theta} = \int_0^\infty \theta E(\theta)\, d\theta = \int_0^\infty \theta\left[-dI(\theta)/d\theta\right] d\theta$$
$$= -\int_1^0 \theta\, dI = \int_0^\infty I(\theta)\, d\theta = 1 \qquad (16)$$

which proves that the mean residence time $\bar{\theta}$ is equal to 1 or that, in ordinary time units, $\bar{t}$ [obtained from a concentration-vs.-time curve at the exit as $\int_0^\infty tc(t)\, dt / \int_0^\infty c(t)\, dt$] is, in fact, equal to V/v, or $\bar{t}_n$. This means, in practice, that the true mean residence time in a vessel can be obtained as the time coordinate of the centroid (center of gravity) of the tracer concentration-vs.-time curve recorded for the vessel outflow.

As has just been shown, this time coordinate will always agree theoretically with the expected mean residence time of the system, irrespective of the nature of the flow in the vessel, as long as correct values for V and v are available.

An age distribution function like $E(\theta)$ or $E(t)$ can be fully described in terms of its moments. For a function $y = f(x)$ the kth moment about the origin, $k = 0,1,2, \ldots$, is defined by

$$M_k' = \left[\int_0^\infty x^k f(x)\, dx\right] \Big/ \left[\int_0^\infty f(x)\, dx\right] \qquad (17)$$

and the kth moment about the mean or centroid μ of the distribution, $k = 0,1,2, \ldots$, is defined by

$$M_k = \left[\int_0^\infty (x-\mu)^k f(x)\, dx\right] \Big/ \left[\int_0^\infty f(x)\, dx\right] \qquad (18)$$

The mean or centroid is itself the first moment about the origin:

$$\mu = M_1' = \left[\int_0^\infty x f(x)\, dx\right] \Big/ \left[\int_0^\infty f(x)\, dx\right] \qquad (19)$$

and serves as a parameter of location of the distribution.

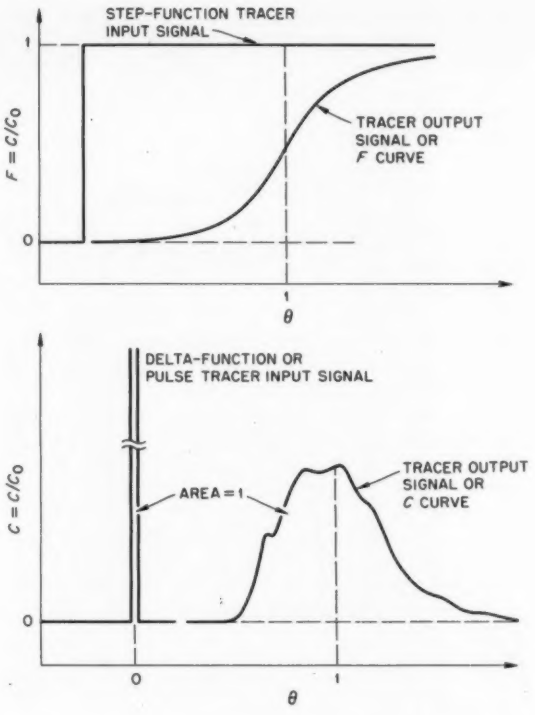

Fig. 2 Response signals of a continuous-flow system, following a step input signal — the $F(\theta)$ function (above) — and a delta-function input signal — the $C(\theta)$ function (below). (After Levenspiel, Ref. 51.)

The second moment about the mean, which is also called the variance, σ^2, measures the spread of the distribution about the mean:

$$\sigma^2 = M_2 = \left[\int_0^\infty (x-\mu)^2 f(x)\, dx\right] \Big/ \left[\int_0^\infty f(x)\, dx\right] \qquad (20)$$

Sometimes higher moments are also found useful — for example, the third moment about the mean, which is a measure of the skewness of the distribution. It is interesting to note that distributions can be compared by studying the corresponding moments without taking the actual distributions into consideration or even knowing the shape of the curves corresponding to the functions $y = f(x)$.

If these concepts are applied to the function $E(\theta)$, one has

$$\mu_\theta = \bar{\theta} = \int_0^\infty \theta E(\theta)\, d\theta \qquad (21)$$

and

$$\sigma_\theta^2 = \int_0^\infty (\theta - 1)^2 E(\theta)\, d\theta \qquad (22)$$

since $\int_0^\infty E(\theta)\, d\theta = 1$.

In terms of the primarily recorded information $c(t)$, we have

$$\bar{t} = \left[\int_0^\infty tc(t)\,dt\right] \Big/ \left[\int_0^\infty c(t)\,dt\right] \tag{23}$$

and

$$\sigma^2 = \left[\int_0^\infty (t-\bar{t})^2 c(t)\,dt\right] \Big/ \left[\int_0^\infty c(t)\,dt\right] = \left\{\left[\int_0^\infty t^2 c(t)\,dt\right] \Big/ \left[\int_0^\infty c(t)\,dt\right]\right\} - \bar{t}^2 \tag{24}$$

The usefulness of these concepts has been demonstrated by Levenspiel and Smith[52] for studying pipe flow, and these authors have indicated a simple way to numerically calculate $\bar{t}$ and σ^2, from which the dispersion coefficient can be calculated if a diffusion-type model is assumed to be applicable to describe the longitudinal mixing.

Danckwerts has introduced the concept of holdback, H, in this context defined as

$$H = \int_0^1 F(\theta)\,d\theta = \int_1^\infty I(\theta)\,d\theta \tag{25}$$

which is the area under the F curve up to $\theta = 1$ or the area under the I curve from $\theta = 1$ to ∞.[50] This means that the holdback represents that fraction of material within the system which has a longer dwell time than the mean residence time. The term H is a useful concept in investigations of chemical reactors since it directly measures the fraction of the reactor volume in which the reaction has proceeded further than desired if the reactor has been designed on the basis of a mean residence time.[53]

Danckwerts distinguishes between two extremes in nonideal steady-state fluid flow through closed systems: either the fluid in the vessel is completely mixed or it is in perfect plug flow. Practical cases are considered as being intermediate between the two. For plug flow the holdback is 0, and for a completely mixed vessel H is $1/e$. For systems possessing large dead-water regions, H approaches 1. When tracer investigations are conducted on continuous-flow systems, using a pulse input, a common result is that $\bar{t}$ as determined from the residence-time distribution will fall below the value V/v. The result may be interpreted to mean that the whole of the volume V is not occupied by the through-flowing fluid; an effective volume V_{eff} can be defined so that $V_{\text{eff}}/V = \bar{t}$ and the difference $V_b = V - V_{\text{eff}}$ is then reported as a blocked or dead-water volume. However, some caution must be exercised in using this concept of blocked volume. There are cases where a low value of V_{eff}/V reveals a fault in the design or operation of the vessel, so that V_{eff} has a direct physical significance; but in systems with a holdback approaching 1, a low V_{eff}/V can be obtained because of the limited sensitivity of tracer detection. A high H value means that the concentration-vs.-time curve [i.e., the $E(\theta)$ curve] has a very long tail that, although it falls below the limit of detection, may give significant contributions to the mean residence time $\bar{t}$, which itself is given by the first moment of the distribution. In this case the whole vessel volume, V, is still physically accessible for the through-flowing fluid, except that the exchange rate of a large fraction of the vessel volume is so low that negligible amounts of tracer have time to enter during the passage of the tracer pulse. Under these circumstances, one should expect that the result obtained would depend on the amount of tracer used; the larger this is, the closer would V_{eff} come to V, and the system would appear to be nonlinear. In spite of this principal difficulty, a tracer test that indicates a low V_{eff}/V is of value from a practical standpoint since vessel characteristics resulting in a flow of this type are, as a rule, not satisfactory for high production of uniform quality. It may be advocated, however, that the recorded concentration curve, whenever possible, should be checked for tracer recovery by setting up a balance; unfortunately the calibrations necessary will complicate the investigation and increase the cost. Another approach to the analysis of blocked volumes or dead-water regions is presented under the discussion of mixed models.

The residence-time distributions, obtained from tracer investigations, usually contain more information than is revealed in calculations of mean residence time and holdback from E or F curves. As a means of extracting more of this information, various mathematical models are used to translate the experimental data into useful sets of parameters that will more fully describe the system being studied.

For systems exhibiting only small deviations from plug flow—e.g., flow through pipes, tubular reactors, and packed beds—the dispersion model (or dispersed plug-flow model) is particularly useful. The fundamental idea of the dispersion model is that the axial mixing can be described by means of a "dispersion coefficient" completely analogous to the molecular diffusion coefficient but acquiring much larger values than this for liquid flow. For this description to be meaningful, the same value of dispersion coefficient, D, will have to be applicable at all points in the vessel. This means, in turn, that the axial mixing is accomplished by a redistribution mechanism which rearranges the fluid elements so many times during their passing through the vessel that the collected effect can be ascribed to a process statistical in nature.

The efficiency of the dispersion process is governed by the dimensionless group, D/uL, called the axial dispersion number, where D is the axial dispersion coefficient, u is the mean linear velocity, and L is the length of the section under study. Conversely, D/uL can be determined from E or F curves in various ways, e.g., as shown by Levenspiel and Smith.[52] An interesting feature of this model is the influence of the boundary conditions at the entrance and exit of the vessel on the relationship between the moments of the C curve on the one hand and the nominal mean residence time V/v and the axial dispersion number D/uL

on the other. This aspect has been discussed by several authors in a series of very enlightening papers.[54–58] If we consider first a test section situated between the point of injection and the (only) point of measurement and have a closed vessel (meaning that no tracer moves into or out of the vessel by dispersion, i.e., $D = 0$ at these points), we will have[55]

$$\bar{t} = \bar{t}_n = V/v \tag{26}$$

and

$$\sigma^2 = 2(D/uL) - 2(D/uL)^2(1 - e^{-uL/D}) \tag{27}$$

Here σ^2 is the relative variance.

If, instead, we imagine an open vessel in which no discontinuity of flow characteristics at the point of injection or measurement exists — L may be a section of an infinite vessel — the corresponding relations are

$$\bar{t} = \bar{t}_n[1 + 2(D/uL)] = (V/v)\,[1 + 2(D/uL)] \tag{28}$$

and

$$\sigma^2 = 2(D/uL) + 8(D/uL)^2 \tag{29}$$

With a vessel closed at one end and open at the other, intermediate relationships are valid in that

$$\bar{t} = \bar{t}_n\,(1 + D/uL) = (V/v)\,(1 + D/uL) \tag{30}$$

and

$$\sigma^2 = 2(D/uL) + 3(D/uL)^2 \tag{31}$$

The smaller D/uL is (i.e., the closer the flow is to plug flow), the more exact will the following approximations for all vessels be, irrespective of termination:

$$\bar{t} = \bar{t}_n = V/v \tag{32}$$

and

$$\sigma^2 = 2(D/uL) \tag{33}$$

The preceding subsection has dealt only with delta-function inputs. Practically, it may be difficult to arrange for a tracer addition that can even approximately be considered to be a delta function. It has been shown, however, that any "single-shot" injection may be used if measurements are taken at two downstream points instead of one. Then if the points of measurement lie between the point of injection and the system exit — the ideal example being an infinite pipe — we have[58]

$$\Delta\bar{t} = \bar{t}_2 - \bar{t}_1 = V/v \tag{34}$$

and

$$\Delta\sigma^2 = 2(D/uL) \tag{35}$$

with

$$\Delta\sigma^2 = \Delta\sigma_t^2/\bar{t}_n^2 = [(\sigma_t^2)_2 - (\sigma_t^2)_1]/\bar{t}_n^2 \tag{36}$$

where length and volume measures refer to the test section. These latter relations indicate a very simple way of conducting and evaluating tests on chemical reactor performance according to the dispersive model.

Also, the case with a time-dependent axial velocity and dispersion coefficient has been treated in the literature. Formally simple relationships are obtained, where time averages of velocity and dispersion coefficients take the place of the constant parameters applicable to steady-state flow conditions.[59,60]

More complex injection techniques have also been used for investigations on closed vessels. Kramers and Alberda[61] have demonstrated the usefulness of the frequency-response method, where the phase shift and the amplitude ratio of the output signal in relation to a sinusoidal tracer input signal are determined. This technique is intimately related to the approach to system dynamics, which is made in control engineering.[62] Some further consideration will be given to this later on in this paper.

When a number of vessels are connected in series, the overall mean residence time of the whole system, $\bar{T}$, will be the sum of the individual mean residence times of the different stages:

$$\bar{T} = \sum_{m=1}^{n} \bar{t}_m \tag{37}$$

and the overall variance

$$\Sigma_t^2 = \sum_{m=1}^{n} (\sigma_t^2)_m \tag{38}$$

since they are means and variances of independent age distributions.

These relations are used in another popular flow model, the tanks-in-series model, which is an alternative way of quantifying residence-time distributions — for packed beds, in particular. In this model the actual vessel is assumed to be subdivided into a large number of perfectly mixed series-connected vessels of equal size. A perfectly mixed vessel is a vessel with perfect longitudinal mixing; nothing is said about the homogeneity of the effluent stream.

For such a model of n stages we have[51]

$$E(\theta) = C(\theta) = \frac{n^n\theta^{n-1}\,e^{-n\theta}}{(n-1)!} \tag{39}$$

with mean

$$\bar{\theta} = 1 \tag{40}$$

and variance

$$\sigma_\theta^2 = 1/n \tag{41}$$

The shape of the E curve of the tanks-in-series model tends toward that of the asymmetric Gaussian distribution of the dispersion model if n is taken large enough, and the models approach equivalence.

The equivalence of the two models for smaller values of n and for larger degrees of dispersion has been discussed in detail by Levenspiel,[63] who identifies variances for the two models, thus obtaining

$$\sigma_\theta^2 = 1/n + 2(D/uL) - 2(D/uL)^2(1 - e^{-uL/D}) \quad (42)$$

However, he points out at the same time that no real equivalence is obtained, inasmuch as the E curves still differ, and that this fact may lead to different conversion yields when applied to chemical reactor calculations. The E curve of the tanks-in-series model can be expressed in terms of the frequency function of the χ^2 distribution, which may facilitate calculations.[64] The tanks-in-series model, in particular, has been favored for describing longitudinal dispersion in flow through packed beds.[13,65] Pippel[66] has suggested a method for finding the effective number of stages of a tanks-in-series model that can be fitted to an experimental residence-time distribution. The problem of backflow between adjacent stages in a tanks-in-series model has been analyzed recently by a number of authors.[67–69]

Treatises on recirculation in technical systems without limitation to particular flow models have been given by Singer, Todd, and Guinn;[70] Metcalfe;[71] and Ljunggren.[72] The same problem has been attacked in a more fundamental way by workers in theoretical biology, for example, Stephenson.[73]

For most vessels used in industrial processes, single-parameter models such as the dispersion and tanks-in-series models fail to account satisfactorily for the through-flow characteristics. The flow in stirred-tank reactors and fluidized-bed reactors, for instance, has to be characterized by more than one parameter, and the through-flow is consequently amenable to analysis only with the aid of multiparameter flow models. The most successful and fruitful approach here is that of mixed models. The real reactor or vessel is subdivided into flow regions—each one being characterized by a simple one-parameter flow model—interconnected to form a composite model whose parameters are adjusted to fit the experimental information recorded in tracer tests on the reactor.

In designing mixed models, elementary flow regions characterized by the following kinds of flow may be useful: plug flow, complete longitudinal mixing, dispersed plug flow, and dead water. In addition, in some cases the following may be required: bypass flow (short circuiting), recycle flow, and cross flow. An early discussion on the use of mixed models to estimate the performance of a real stirred-tank reactor has been published by Cholette and Cloutier.[74] The best fit was obtained by combining a region of longitudinal mixing with one of dead water and allowing for some bypass of fluid. A dependence of the model parameters on the agitation rate could be demonstrated.

A more general treatise on the use of mixed models has been given by Levenspiel,[51,75] who also discusses the definition of dead-water regions. Arguing that any material spending a time longer than, say, $2\bar{t}_n$ in the reactor will contribute negligibly to its performance, he suggests that a cutoff be made at this point in the residence-time curve and the "active" flow rate and the corresponding "active" volume (which is the complement to the dead-water volume) be calculated from the effective mean residence time computed from the truncated curve. Levenspiel also discusses at some length the application of mixed models to flow in fluidized-bed reactors.[51]

If the model is made sufficiently complex, it can be adapted to cover any given sets of experimental data, but it may still fail to predict the result of new experiments on the same system. In general, the most sensible results will be obtained with the simplest model that will account for the data at hand reasonably well and whose compartments are suggested by the actual system. The descriptive mean ages for the combined system can then be found from those of the various regions in the following manner:

The mean age of the fluid in the exit stream from series-connected regions 1,2, ... is

$$\bar{t} = \bar{t}_1 + \bar{t}_2 + \ldots \quad (43)$$

while for regions in parallel

$$\bar{t} = (V_1 + V_2 + \ldots)/v \quad (44)$$

where V_1, V_2, ... refer to the active volumes of the system, and v is the through flow.

Some recent approaches to mixed-model representation have been made. Wolf and Resnick[76] assume an $F(t)$ function of the form

$$F(t) = 1 - \exp\left(-\eta\,\frac{t-\epsilon}{\theta}\right) \quad (45)$$

to be valid for $t \geq \epsilon$, while

$$F(t) = 0 \text{ for } 0 < t \leq \epsilon$$

A straight line will be obtained if $\ln(1-F)$ is plotted vs. t/θ. From such a plot the two parameters η and ϵ may be determined. This method can be used for both single and multistage systems to account for deviations from ideal plug flow or perfect mixing, caused by dead space, short circuiting, error in average residence-time determination, lag in response, or any combination of models.

Another approach has been taken by Naor and Shinnar[77] who introduce a function $\lambda(t)$, which is

analogous to the intensity function of statistical mathematics. The function $\lambda(t)\,dt$ denotes the probability for a particle that has been in the system for a period, t, to leave it within the next time element, dt. The following relationship holds:

$$\lambda(t) = -d[\ln I(t)]/dt \tag{46}$$

$I(t)$ being the internal age distribution. The authors claim that this mode of representation enhances the interpretability of experimental curves. The fact that transfer characteristics of mixed models can easily be expressed in terms of the transfer functions of the individual regions is a great advantage. For calculations of this type, Laplace transformations are most useful.

The theory of tracer experiments, as far as it has been discussed up to now, has been confined to the flow of fluids under physically well-defined conditions, without any consideration of chemical transformations. We have actually thought of the tracer as a substance that is carried through the vessel by the stream of fluid and whose behavior can in no respect be distinguished from that of the fluid, with the exception of the property that makes it detectable. Bergner's[46] terminology suggests that the mother substance of the tracer has to be the fluid itself and not any of the process chemicals carried by the fluid if knowledge of the transfer characteristics is to be obtained which can be of general applicability in calculating conversions, etc.

Conversely, process chemicals that undergo conversion in the system can be added in radioactive form to gain information on the specific fate encountered by those particular compounds. But when chemical changes are involved, there exists a much more difficult problem than the mere physical transportation of tracer and mother substance. We now want to follow the individual atomic species, and our system or compartment may have to be defined chemically instead of geometrically, or both. This is also the stage where radioisotope tracers, by necessity, must be chosen. Problems of this type are encountered in biology, medicine, and drug technology, and the main efforts in developing a general tracer theory were made in the field of theoretical biology. A recent review by Bergner[46] gives an excellent analysis of the fundamental concepts of tracer theories and also looks at the evolution of present-day theories in a historical perspective.

The first one to use a model for a metabolic system, consisting of a number of well-stirred compartments between which the substance was exchanged according to first-order kinetics, seems to have been Teorell[78] in 1937. Apart from this, many intuitive methods have been employed in the biomedical field for evaluation of tracer experiments using semilogarithmic presentation of tracer concentration-vs.-time plots to establish measures like the "last linear slope" which were then evaluated—on the basis of a concept of isotopic equilibrium due to complete mixing after sufficient time—to give relations between mean residence times, pools (volumes), and flow rates. A classical paper by Sheppard and Householder[79] in 1951 was the introduction to the more fundamental theoretical method of compartmental analysis. Further contributions were given by Berman and Schoenfeld[80] in 1956, and a fuller account was later given by Sheppard.[81] Compartmental analysis has acquired a place as the most commonly used method today for analysis of tracer data.

Historically, the use of tracer methods in biomedicine started with the work of Stewart[82] in 1897, later continued by Hamilton,[83] for measuring flow rates and volumes of blood. As a result of their work, a set of equations, generally referred to as the Stewart–Hamilton equations, was established and may be written

$$Q = v \int_0^\infty c(t)\,dt \tag{47}$$

and

$$V = \frac{v^2}{Q} \int_0^\infty tc(t)\,dt = v\,\frac{\int_0^\infty tc(t)\,dt}{\int_0^\infty c(t)\,dt} \tag{48}$$

where the symbols have the same meaning as before. The first of these equations may be considered to be a simple material balance for the tracer and the relationship between the second one (Eq. 48) and Eq. 16 is evident. A derivation of these equations in statistical terms was given by Meier and Zierler in 1954 (Ref. 84).

The "total-count equation" (Eq. 3) states the same fact as Eq. 47 in terms of the digital measurement principle specific to this method. There have been different views expressed regarding the identity between these two representations.[23,85–87]

A recent contribution to the theory of tracer methods has been given by Bergner who has discussed the fundamental aspects of the tracer principle and, in a new set of concepts called "the tracer dynamics," has built a rigorous theory of general validity.[88–91]

Bergner's central formula is

$$b^\circ = \xi^\circ_{Se} \int_0^\infty \frac{b(t)}{b_s(0)}\,dt \tag{49}$$

which is valid for any compartment. Here b° is the total amount of mother substance in the compartment, i.e., total volume or pool size; ξ°_{Se} is the total turnover, i.e., total flux of mother substance from the environment into the compartment (which equals ξ°_{eS}, the flux of mother substance from the compartment to its environment, since the mother substance is assumed to be in steady state); $b(t)$ is the amount of tracer in the compartment at time t; and $b_s(0)$ is the

total amount of tracer supplied to the compartment at time $t = 0$. If we make the following identifications:

$$b^\circ = V$$

$$\xi^\circ_{Se} = v$$

$$\frac{b(t)}{b_s(0)} = I(t) = \frac{v}{V} I(\theta) \tag{50}$$

$$\frac{b(t)}{b^\circ} = c(t)$$

and

$$b_s(0) = Q \tag{51}$$

the similarity between Bergner's results and those given in Eqs. 9 to 16 is evident. Noting that b and $b_s(0)$ can be interchanged in Eq. 49, we can see that the Stewart–Hamilton equations (Eqs. 47 and 48) are contained in this formula. Bergner also discusses in detail which restrictions must be placed on the injection for the relationship in Eq. 49 to be valid. The shape of the equation underlines an important fact: We have to observe the whole tracer retention curve $b(t)$ and cannot, without additional hypotheses, draw conclusions from isolated sections of the curve.

The residence-time distribution describes the overall properties of a continuous-flow system, but it does not reveal where the actual flow paths are located within the system. The existence of various flow regions (cf. Mixed Models) and of special deficiencies such as channeling can be verified from the residence-time distribution, but the underlying causes, structural or other, cannot be brought to light. A mapping of flow patterns inside the system is necessary to achieve this. Some knowledge of the internal flow patterns can be gained by external measurements for systems that have more than a single inlet and outlet, since, in principle, tracer experiments can be arranged to determine tracer carryover and residence-time distributions by injecting tracer into one inlet at a time and recording tracer concentration at each outlet as a function of time. In this way, residence-time distributions pertaining to any given pair of inlets and outlets can be established, as can the branching of tracer (and process chemicals introduced in the same way) from the inlet to each one of the outlets; a tracer balance can be established by means of Eq. 47 if the flow rates at all exits are known.

In a qualitative fashion it is also possible to gain insight into the local flow patterns of a system of not too large dimensions by placing external detectors at different points close to the vessel walls and recording the intensity variation at these points after introduction of a gamma-emitting tracer into the system inlet.

For a detailed elucidation of flow patterns, however, it is necessary to arrange measurements inside the system, either by placing detectors at or by sampling at various points in order to obtain local concentration-vs.-time relationships. Also, this aspect has been treated theoretically by Danckwerts,[92] who points out that the average age of material at any given point inside a process vessel can be found as

$$\bar{t} = \frac{v}{Q} \int_0^\infty t c(t)\, dt \tag{52}$$

following a sudden injection of the quantity of tracer Q at the inlet. This average local age is a measure of local rate of replacement and can thus be expected to indicate the possible existence of stagnant or bypass zones. One would expect the integral to assume values approaching zero near the system inlet and tending to approach the value of the average age of material in the outgoing stream near the exit. By drawing contour maps of average (or median, whichever may be simpler) ages over the volume of the system, one can readily show areas of stagnancy or channeling.

INVESTIGATIONS OF INDUSTRIAL-PLANT SYSTEMS

Flow of Fluids. Information obtained in tracer investigations characterizes continuous-flow systems as to their "concentration dynamics" using a term suggested by Gilath.[93] Valuable knowledge of the steady-state and transient behavior of a system can be derived from such results, which may lead to a better understanding of the physical processes taking place in the system and a check on the effectiveness of practical designs. When the residence-time distribution of the fluid particles in the through-flowing stream has been established, the conversion yield of any given first-order chemical reaction can be directly calculated and bounds for conversions of nonlinear reactions can be set.[51,53,94,95] It is also possible to calculate the response of the system to disturbances of various kinds and to estimate quantitatively the degree of quality uniformity for systems where the process requires a minimum of spread in residence times, i.e., plug flow. The existence of very long turnover times in stagnant zones can be revealed and the risk assessed for adverse phenomena such as unwanted condensations or deposits.

Among the systems investigated are (1) those in which the transported medium is a fluid (or at least fluidized): pipes, continuous blenders, pulp-bleaching towers, digesters, contactors, heat exchangers, holding tanks, packed beds, fluidized beds, flotation cells, catalytic crackers, stills, and glass furnaces; and (2) those in which solids are conveyed: rotary kilns, dryers, furnaces, and digesters.

The dispersion in pipe flow, as mentioned previously, can be studied conveniently by using radioisotope tracers. The results that were obtained either directly or by calculations can be employed, by use

of dispersion indexes established in measurements, for assessing the intermixing between two fluids transported consecutively through the same pipeline. The intermixing determines the degree of contamination that occurs at the interface.[14,96]

Mixing in continuous blenders is a process that readily lends itself to control using radioactive tracer added in sudden injections to the system inlet. Emphasizing the usefulness of the pulse testing method, Gardner et al.[97] investigated an aqueous mixing system containing three series-connected tanks using $^{113m}InCl_3$ as a tracer. The ^{113m}In was obtained by milking a ^{113}Sn source.

A large-scale test has been carried out by Stemerding et al.[98] on a stirred vessel used for contacting a hydrocarbon and an aqueous stream. Simultaneous tracing of the hydrocarbon phase with ^{14}C-labeled *n*-decane and of the aqueous phase with $Na^{131}I$ was performed.

In petroleum refineries an impressive number of tracer investigations have been carried out. Mixing in surge tanks and stills has been checked by Hull et al.[99] using a chelate compound of ^{95}Zr and ^{60}Co naphthenate, respectively, as oil-soluble tracers. The study is a practical demonstration of the use of Danckwert's concepts of scale of segregation and degree of segregation to describe the performance of continuous blenders.[50] General surveys of the use of tracers in refinery control have been given by Hull,[100] King,[101] and, very recently, by Mott.[28] King[101] describes a test of the relative efficiencies of an old jet-injection device and a modern jet tray, with respect to back mixing in an olefin alkylation plant.

Ausman et al.[102] have used ground firebrick impregnated with ^{24}Na to evaluate the mixing performance of a fluid coker.

Much interest has been devoted to studies of flow patterns, mixing, and loss of catalyst material in fluidized catalytic cracking units. Essential information has been obtained using tracer investigations on the following:[28] (1) initial loss of fresh catalyst after addition to a unit; (2) mixing pattern and mixing rate of fresh catalyst with the catalyst material in a unit; and (3) the long-time loss of catalyst from a unit. Tagged catalyst is injected into the unit, and the activity variation subsequently is followed from outside the unit or from samples taken at appropriate points. Such studies have been published by Hull,[103] Hull and Ball,[104] Singer, Todd, and Guinn,[70] Guinn,[105] Kinsella and Mitchell,[106] Hull and von Rosenberg,[107] and Todd and Wilson.[108] Counting techniques for such studies have been described by Guinn, Lukens, and Wagner.[109]

Handlos, Kunstman, and Schissler[110] have investigated the gas-mixing characteristics of a fluid-bed regenerator. They used helium as a tracer but point out that the large-volume injections which had to be made in some cases changed significantly the actual gas flow within the system. The use of ^{85}Kr, according to the authors, would have eliminated this disturbance.

Fractionation in refining processes can be investigated to advantage by addition of radioactive tracers and subsequent measurement of activity in a product stream. Fries, Hull, and Dupzyk[111] used hydrocarbons labeled with ^{14}C or ^{3}H as tracers and a flow liquid scintillation counter that monitored a continuously drawn sample from the relevant product stream. By applying the total-count principle, one can determine what fraction of a given, labeled compound comes out each product line.

Polish workers have developed a range of organometallic compounds based on germanium (^{77}Ge) from which a useful tracer can be selected for almost any given hydrocarbon fraction.[112,113] Mott,[28] on the other hand, emphasizes the general usefulness of bromobenzene (with ^{82}Br).

Flow patterns in liquid systems have also been studied; the flow distribution across the trays in a large vacuum distillation column has been mapped using triphenylstibine (^{124}Sb) (Ref. 114).

Provision for high quality of the distillate from refinery vacuum-distillation units requires a low degree of contamination with nonvolatile feed constituents, and therefore entrainment within the unit has to be kept small. With the use of $^{140}Ba-^{140}La$ as a tracer in the form of a phenolate and the monitoring of the activities of distillate, pitch, and runback line, the magnitude of the entrainment could be estimated by considering the tracer balance.[115] Similarly, entrainment in an alkylation plant has been measured.[32]

Very short residence times have been studied in a sulfuric acid alkylation system containing a circulation pump, a heat exchanger, and a tank.[116] Barium-137*m* (half-life 2.6 min) milked from a ^{137}Cs column was used as a tracer.

An extensive study of the flow characteristics in a fluidized-bed reactor has been performed by May[117] who used ^{132}I impregnated within the solid catalyst material and helium gas as tracers. Measurements were made of gas flow rates, of gas volumes in bubbles and emulsion, of mass transfer between bubbles and emulsion to obtain the "cross-flow," and of catalyst mixing to establish dispersion coefficients.

Yavorsky and Gorin[118] used irradiated coal particles of various sizes to study movement and disposition in a fluidized-bed reactor. Both the rate of segregation of various-size coal particles and their equilibrium disposition could be studied using an external scintillation counter.

DeMaria and Longfield[119] have attacked the problem of determining local residence times in the gas phase of fluidized beds.

One large area of tracer application is the study of liquid or gaseous flow through packed beds and columns of various kinds. Markas and Beckman[120] used iodinated triolein (^{131}I) to label the dispersed toluene phase in a packed countercurrent liquid–

liquid extraction tower. The main purpose of the investigation was to measure the holdup characteristics (fractional volume of the dispersed liquid phase) when different sizes of Raschig rings were used as packings, but conclusions concerning channeling could also be drawn.

Sater and Levenspiel[121] have measured the axial dispersion coefficients in both gas and liquid phases in a bed packed with Raschig rings or Berl saddles. In the gas phase ^{41}Ar was used as a tracer, while $Na^{131}I$ served the same purpose in the liquid. Empirical relations were obtained for the dispersion coefficients in terms of Reynolds number, axial flow velocity, and size of packing.

Another example of investigations on the liquid flow through a packed bed can be taken from the cellulose industry, where the circulation through two types of sulfite digesters for pulp making was studied by means of repeated residence-time measurements for the circulating cooking liquor during the digesting.[122] It became evident that the flow rate decreased toward the end of the cooking and also that one of the digester constructions (with a bottom countercurrent liquor circulation) investigated was superior to the other since in this one the variance of the residence-time distribution was much smaller at the end of the cooking. The digesters are schematically shown in Fig. 3, and the variance of the residence-time distribution as a function of time is given in Fig. 4. Similar studies on liquor flow in continuous digesters have also been made.[123]

The pulp and paper industry has made much use of radioactive tracers to test the efficiency of various process units and to reveal deficiencies affecting the production capacity or the quality of the product. In the great majority of reported investigations, the attention is focused on the flow of the cellulose fiber itself rather than on the aqueous phase carrying the suspended fiber. Therefore the tracer used must have properties identical with those of the natural fiber. One way to achieve this is to tag the cellulose fiber by incorporating a suitable radioactive substance. Allen, Rezanowich, and Mason[124] incorporated ^{131}I into the fiber by first precipitating silver chloride inside the fiber and then letting the chloride exchange with radioactive iodide. This fiber has been used for investigating fiber flow in paper-making machines when comparing differently designed head boxes[125,126] and for evaluating centricleaner performance at different pulp consistencies.[127] The making of the labeled fiber according to this procedure is, however, a tedious operation and this tracer, for most investigations, has now been abandoned in favor of glass fiber cut in lengths to resemble those of cellulose fiber. The glass fiber, upon neutron irradiation, has sufficient ^{24}Na activity to serve as a most useful and readily producible tracer. The larger specific gravity of the glass compared with that of the cellulose has not been observed to cause any nonrepresentative behavior above concentrations of 0.5%.

The investigation of the flow in bleaching towers for pulp and in chests intended for mixing or for storing of pulp has become an established routine.[123,128] A bleaching tower is an excellent example of a unit that should possess plug-flow characteristics, in

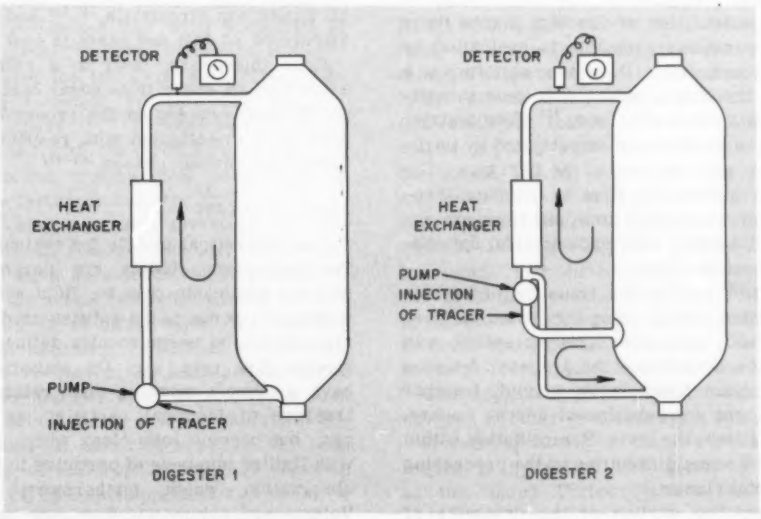

Fig. 3 Schematic drawings of two sulfite digesters, demonstrating the different circulation systems and the measuring arrangement in the tracer investigation. (After Burchardt, Ljunggren, and Stockman, Ref. 122.)

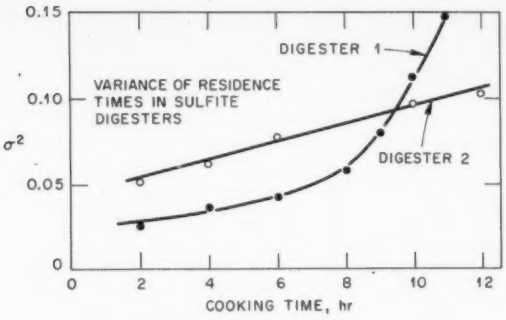

Fig. 4 Variance of the residence-time distributions vs. cooking time for digesters 1 and 2 of Fig. 3. The sharp increase toward the end of the cook for digester 1 is noticeable; this corresponds to a proportional increase in the dispersion coefficient (Eq. 33). (After Burchardt, Ljunggren, and Stockman, Ref. 122.)

contrast to most of the investigated chests that should accomplish an efficient mixing of various qualities of pulp entering the chest. For the sake of constancy in product quality, it is also essential that a mixing chest be capable of smoothing concentration variations that may appear in the feed streams. Ryti[129] has made a systematic study of the ability of pulp mixing vessels of different construction to attenuate disturbances of various duration appearing at the inlet.

Gilath et al.[130] have studied the mixing in a pulp chest. Using the mixed model approach of Wolf and Resnick,[76] i.e., plotting $\ln(1-F)$ as a function of t/θ, they were able to state that 83% of the chest volume gave perfect mixing, 12% represented dead volume, and 5% represented plug flow.

In mineral-concentration or leaching plants, there are numerous operations amenable to evaluation by means of tracer techniques. Dorr-type agitators in a pilot plant for leaching uranium ore were investigated as to their mixing efficiency.[131] Four agitator tanks connected in series were investigated by introduction of tracer into the feed of the first tank. The investigation is one of the first to compare theoretically derived residence-time distributions for series-connected mixers with experimental data obtained with radioactive tracer.

Eichholz et al.[132] studied the transport of the material in a flotation circuit using irradiated zinc ore as well as $^{64}CuSO_4$ as tracer (copper sulfate was used as a surface activator in the process). Various retention and contact times in the circuit, together with figures on the consumption of copper sulfate, could be derived from the tests. Recirculation within the circuit caused some difficulties in the processing of the experimental curves.

More recently, two studies of the dynamics of flotation cells have been reported by Niemi.[133,134] The mass transportation both in mechanically agitated and in pneumatic flotation cells was studied using $^{24}Na_2CO_3$ as a tracer for the fluid, irradiated tailings (producing ^{24}Na as the dominating activity at the time of measurement) for the gangue components, and irradiated chalcopyrite for the floatable components. The experimental results were fitted to theoretically derived models, and it could be shown that the residence-time distribution of the pulp body in the mechanical cell corresponds to that of an ideal mixer in series with a small plug-flow region. The tracer investigations permitted the establishment of the essential transfer functions of the system. It deserves mention that, in the evaluation, the residence-time distributions obtained experimentally required correction for recirculation.

An interesting study has been made at a nuclear reactor installation into the function of a delay tank intended to hold the circulating water long enough for the induced ^{16}N activity to decay to a harmless level before the water entered the heat exchangers.[135] Considerable axial mixing was found to exist which would have led to a prohibitively high ^{16}N-activity level, but the error could be eliminated by reconstruction.

Several investigations have been made of flow patterns and residence times in glass furnaces.[136–139] Several different tracers were used: ^{32}P incorporated in glass, tantalum oxide (as inactive tracer), ^{46}Sc incorporated in glass, and most frequently, ^{24}Na induced by irradiation of actual glass taken from the process.

Radioactive gas tracers—^{85}Kr, ^{41}Ar, ^{133}Xe, and ^{82}Br-labeled methyl bromide—have been used for various investigations of air turnover and flow patterns in connection with ventilation problems in houses,[140] with the drying of hay in haylofts by means of forced air circulation,[141,142] and with mixing and dispersion of dust and gases in coal mines.[143,144]

Dorr thickeners, used in a refining step in the extraction of sugar from sugar beet and in the Bayer purification process in the production of aluminum, have been investigated with respect to the throughflow of the various phases.[145–147]

FLOW OF SOLIDS

The theoretical models for evaluating results from residence-time studies are derived for, and are strictly applicable only to, fluid systems. For solid materials, some of the entities used become obscure and cannot be unequivocally defined: volume, volumetric flow rate, etc. On statistical grounds they have a simple meaning when related to very large numbers of identical particles, as in a liquid or a gas, but become less clear when one is confronted with limited numbers of particles in the volume under observation, which, furthermore, vary with size. Volume and volumetric flow rate more or less lose their meanings for solid particles; instead, mass and mass per unit time must be used. In line with the different phenomenology, an entirely different situa-

tion arises when solids are to be labeled radioactively. The labeling has to be done judiciously so that the tracer material is truly representative of either the average properties of a nonhomogeneous material or of the properties of some especially interesting fraction of it.[1] It is noteworthy, however, that practical useful information can also be obtained from investigations where the question of defining the tracer material has been treated rather lightly. The following methods to accomplish labeling of solids have been used:

1. Irradiation of the actual process material to induce useful activities
2. Affixing a radioactive substance to the surface of the process material by adsorption or impregnation
3. Mechanical tagging with a radioactive source
4. Making a physical limitation of the material under study which is easily rendered radioactive

The first two methods are generally used when a large portion of a fine-grain material is to be labeled for obtaining statistically valid results, while the third and sometimes the fourth alternatives are used when individual large lumps are to be followed.

As early as in 1951, coal travel in continuous vertical retorts was studied using small ^{60}Co sources.[148] Measurements of the rate of descent of the charge in blast furnaces and other furnaces have been reported by many authors.[29,149–152] A similar study of the movement of iron-ore pellets in a Wiberg–Söderfors sponge-iron furnace was made in conjunction with measurements on the distribution of reductive gas flow across the furnace.[153]

The material transport in rotary kilns used in many industrial processes has been subjected to intense study by means of radioactive tracers. Rutle,[154] in 1955, investigated the transport in a rotary kiln for making portland cement, and this study was followed by several others.[153,155,156] Some investigations were also concerned with the circulation of alkali in the kiln.[153,157] Similar studies have been made on drying kilns in the fertilizer industry.[158,159]

In Poland[160,161] very extensive investigations into material transport in rotary kilns for various industrial purposes have been made, which emphasize that knowledge of the actual residence time in various zones of the kiln is of importance, not only for choosing optimum operating conditions for existing kilns but also for reconstructing kilns and designing new units. Kilns for the following purposes have been investigated: (1) decomposing calcium sulfate to SO_2 and cement clinker; (2) sintering apatites with soda during production of thermophosphate; (3) reducing acidic iron ores; (4) fire enrichment of zinc ores; (5) calcining soda in rotary kilns; and (6) producing cement clinker.

In the cellulose industry, continuously operating digesters have been introduced on a large scale in the last decade or so, and more than half of all the paper pulp in the world is now being made in continuous units. In introducing these digesters, radioactive tracers played an important part because they made studies of the chip transport in the digester possible. It is absolutely essential to ensure that all chips are transported through the digester at the same speed irrespective of their positions in the cross section in order that the uniform cooking necessary for the production of high-quality pulp should be obtained. Fineman[162] has described a method where pieces of radioactive copper wire were inserted in actual wood chips. Because of the radiation characteristics of ^{64}Cu, the method is usable only for small-to-medium-sized digesters, and alternative techniques using ^{140}La "baked" into plastic chips[163] or ^{24}Na enclosed in small Teflon containers[123] have been developed and used on digesters of up to 4.3 m in diameter.

Diamond imitations, made of an Al–Co alloy of exactly the same specific gravity as diamond, have been used to check on the operational efficiency of diamond concentrators.[164]

Investigations of Natural Systems

There is a far-reaching analogy between tracer investigations of material transport performed in industrial plants and in natural systems as far as the principal arrangements of the investigations and the evaluation of the results are concerned. Similar transportation mechanisms act on tracer and traced substance in the plant and in the water, ground, or atmosphere around us. There is usually a difference in scale, however. While plant investigations are limited as to the size of the studied systems—and thus the amounts of tracer required—natural systems are not confined in the same way. Sooner or later, dilutions are reached which lower the concentration of tracer below the limit of detection unless copious amounts of tracer are used, as in the global tracing of tritium of atmospheric origin and from nuclear weapons tests. The applications of radioactive tracers to natural systems extend over wide areas in geology and geophysics, soil science, hydrology, and meteorology. It is neither possible nor is it the intention to give a full review of these applications in this paper; the account will be limited to some methods and investigations that are related to the in-plant applications described earlier.

The applications in geology and geophysics are covered in other reviews at this symposium.[165,166]

A general survey of the use of radioisotope techniques in hydrology was given by Payne et al.[167] at the Third United Nations Geneva Conference, 1964, and a more recent report was published after a Working Group meeting in October 1965 (Ref. 42).

In hydrology, two fields of particular interest can be distinguished: (1) studies of groundwater flow and

(2) determination of flow patterns and dilutions of discharges into open-water receivers.

The role of isotope methods in groundwater research has been discussed by Harpaz et al.[168] Among the uses found for radioactive tracers are:

1. Determination of the physical characteristics of aquifers (permeability, porosity) from flow rate measurements of injected tracer pulses
2. Measurement of dispersivity
3. Studies of the chemical properties of the medium
4. Determination of flow direction and velocity, both local and averaged over a recharge area
5. Studies of mixing and dispersion patterns

In many of these applications, radioactive tracers are a unique tool. A report on various flow models for evaluating sudden-injection tracer experiments in aquifers has been made by Halevy and Nir[169] who also introduced the concept of transform function of an aquifer.

An interesting case study is the investigation reported by Burdon et al.[170] of underground interconnections between sinkholes and springs in a karstic limestone region in the Greek Peloponnese, and the subsequent analysis of the recorded tracer concentration curves by Clayton and Webb[171] with a view to obtaining quantitative information on the nature of the underground stream.

Water pollution is a formidable problem today in many areas of the world because of the discharge of ever increasing volumes of effluent from industries and communities. Radioactive tracer methods, at an early stage, were used as an aid to determine flow patterns and dilutions from existing or planned effluent outlets.[172–174] An important generalization of the earlier methods, especially suited for prediction of sewage dilution from planned outlets, was presented by Berg and Somer[175] in 1962. The method is based on integrating the tracer concentration-vs.-time curve recorded at fixed points following a sudden release of a known amount of tracer at the point where the future outlet is foreseen. In this way, dilution figures applicable to the conditions of a continuous discharge at the chosen point are obtained. This means that several suggested sites for a planned outlet can be investigated as to their ability to disperse effluent without first making costly installations. Harremoes[176] has reported on several such investigations and on the experimental technique used; he has also described a further development in the applications of these methods to outlets located at the bottom, in cases of jet diffusion and stratified dispersion.[177] These methods have been used on a large scale in a joint operation between the Danish Isotope Center and the Isotope Techniques Laboratory to analyze the conditions for sewage disposal in the northern region of the sound between Sweden and Denmark.[178]

A recent advancement in measuring techniques has been made with the thermoluminescence dosimeters which, when made of calcium sulfate or calcium fluoride, can measure doses well below 100 μr. As has been demonstrated by Bjärngard,[179] such dosimeters can be placed at points in the vicinity of a sudden or extended release of active material to measure, after calibration, the integral of concentration with respect to time. They are usable in air as well as in water, although the limit of detection will be at a higher concentration in air because of the higher background. A pattern of isoexposure curves resulting from a release of 0.2 curie of ^{82}Br is shown in Fig. 5.

A special instrument to measure the precise direction and flow of slow-moving currents especially at great depths has been developed to serve oceanographers and hydrologists.[180] The DWICA, or deep-water isotopic current analyzer, operates by injecting ^{131}I in solution from the center of a circular array of counting tubes. Drift direction and velocity of the released tracer yield direction and velocity of the water flow.

Closely related to the use of radioactive tracers in hydrological problems is the use of tracers to follow the movement of sand, sediment, and riverbed loads. These applications have been reviewed by Griesseier[181] and fundamental aspects discussed by Crickmore and Lean.[182] Some recent information has been collected at the Working Group meeting on isotopes in hydrology in 1965 (Ref. 42) in a published discussion[183] on measurement principles.

Air movements in clear air and clouds can be mapped by releasing a suitable tracer from an aircraft and then measuring the distribution with a detector located in the aircraft. Such studies are of importance in connection with research on cloud seeding for increasing precipitation in arid zones. These studies have been performed using copper oxide (^{64}Cu) for a tracer.[184]

Studies of the movement of air and air pollutants from industrial or community installations is of considerable importance. All risks of spreading radioactive materials over large areas can be avoided in making these studies by the use of inactive tracer substances, e.g., antimony oxide, in combination with subsequent activation analysis.[185,186] The use of quasi-stable ^{129}I as a tracer for this purpose has been suggested.[187,188]

Development Trends

The wide variety of uses of radioactive tracers for studying the flow of materials and measuring the parameters of flow has been demonstrated in the review. One might ask if primarily new uses of radioactive tracers are to be expected in this context. It may be stated that the only limitation to the development of new ideas in tracing lies in the human imagination. This statement is, of course, not strictly true, but it emphasizes the extremely wide ap-

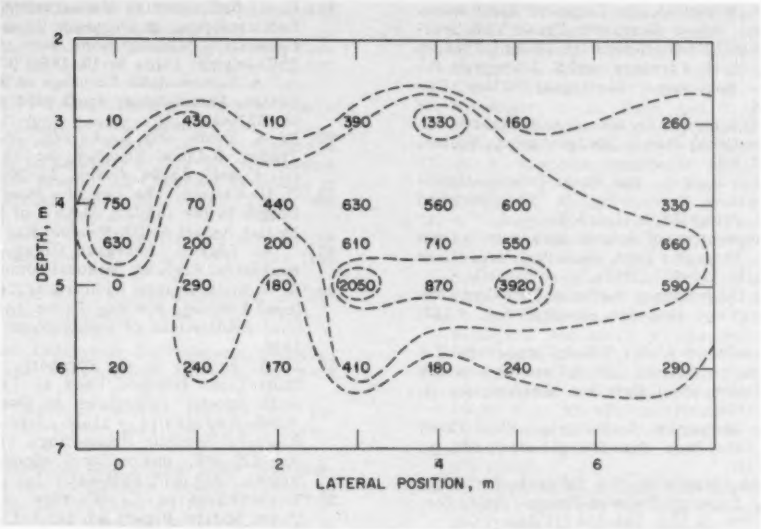

Fig. 5 Exposure pattern following a release of 0.2 curie of ^{82}Br in water registered by means of thermoluminescence dosimeters. Exposure is given in μr, and isoexposure lines have been drawn for 200, 400, 800, and 1600 μr. The distance from the release point to the plane of measurement was 10 m. (After Björngard, Ref. 179.)

plicability of the radioactive tracer method. If one were to point to probable areas of extended use in the near future, the most important area seems to be that of process control. There are two aspects of the use of tracers for this purpose:

1. Tracer investigations are used during the development of a computer control system, together with other methods, as a means of analyzing the process for obtaining transfer functions and other pertinent information.

2. Intrinsic radioactive tracers are used in the process as a link in a control system to provide a signal proportional to the concentration of some vital component or for some similar purpose.

The first alternative, which integrates the tracer methods into modern systems engineering, has been discussed by Rotkirch, Kommonen, and Castren.[189] These authors give prominence to mathematical studies and mathematical understanding of the actual process, e.g., by computer simulations, and suggest that tracer methods be used restrictively to provide specific data only when other methods cannot compete. There seems to be little doubt, however, that the tracer method using radioactive substances is outstanding in providing data on absolute or relative transfer functions and for applying the pulse testing principle under full-scale operating conditions in industrial plant as well as in pilot-plant runs.

The use of intrinsic radioactive tracers for process control, although not used under plant conditions to any extent today, opens up interesting possibilities for controlling various unit operations by continuously feeding radioactive tracer into the process stream.[190] Feasibility studies have been made of many actual industrial processes that involve, for instance, the following of a major component during processing or of the removal of a critical impurity from the major product.[191] Experimental studies with tracers have been made on the removal of impurity metals such as copper and iron in electrolytic refining of nickel.

Instead of injecting radioactive tracers into the process stream, they can alternatively be produced inside the stream by irradiation with an external neutron source. In this way a control system may be developed which works as a continuous on-line activation analysis.[192]

Unquestionably, more development work and more trials in industrial plant will be needed to establish radioactive tracers as important information sources in designing control systems or as permanent links in such systems, but, if these efforts are made, it is also certain that radioactive tracers will contribute largely to the continual automation of the process industries. (FJM)

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RADIOISOTOPE THICKNESS GAGE FOR HOT-STRIP MILLS

A thickness gage based on attenuation of ^{137}Cs gamma radiation is said by its manufacturer, Hitachi, Ltd., of Tokyo to be "the world's first thickness gage, for *hot*-strip mills, to use radioactive isotopes." The gage measures thicknesses, from 4 to 50 mm, of hot-rolled steel plate at temperatures of 600 to 1200°C without making any contact with the steel. The accuracy is ±0.5% of the thickness or 0.08 mm, whichever is greater. The source is installed at the top of a 4-m-tall C frame and the detector at the bottom. The hot-rolled steel plates pass between the source and detector. Thickness is recorded on the indicating instrument panel. The desired limits are set on the operating panel, and the recorder stops if the limits are exceeded. (From Type RTI-S Gamma-Ray Thickness Gauge, *Hitachi Review*, 16: 135 (1967). (MG)

Section II

Isotopes and Radiation Technology

Food Irradiation

Bibliography Covering Irradiation of Fruits and Vegetables

A. S. Abdel-Kader and E. C. Maxie of the Department of Pomology, Davis Campus, University of California, have compiled a bibliography of more than 500 references, through 1965, dealing with various aspects of irradiation of fruits and vegetables. The references have been supplemented by an author and subject index and are available as Report ORNL-IIC-11 free of charge from the Isotopes Information Center, Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830.

The bibliography includes approximately 260 references of a general nature, pertaining to the overall problems, advantages, disadvantages, and potentials of fruit and vegetable irradiation, with 30 more citations dealing specifically with fruit and 4 more with vegetables. Other subject headings (with the approximate number of references in parentheses) are as follows: Containers and Packaging (6); Dosage (3); Economics (13); Foreign Work (115); Fruits and Nuts: Apples (15); Bananas (2); Cherries (1); Chestnuts (4); Citrus (12); Figs (1); Grapes (8); Mangoes (2); Peaches (5); Pears (3); Persimmons (1); Prunes (1); Strawberries (19); Whortleberries (1); Irradiator Design (14); Physiological and Biochemical Effects (100); Quality and Texture (35); Safety (3); Sources (3); Effect on Specific Organisms (11); Storage (7); Vegetables: Artichokes (3); Asparagus (1); Beans (5); Beets (2); Broccoli (4); Carrots (13); Corn (1); Onions and Garlic (14); Potatoes (75); Radishes (1); Sweet Potatoes (2); Tomatoes (9).

Hopefully this material will be updated and perhaps broadened to include other phases of the food-irradiation program. (P. S. Baker)

Replacement of Underwater Bearings in a High Radiation Field

By John B. Huff and Joseph M. Lee*

Abstract: A procedure and tools for replacing underwater bearings were developed and were used to replace worn ball bearings in the MPDI source-elevator mechanism with solid bearings.

The Marine Products Development Irradiator (MPDI),† which is being used to study radiation pasteurization of fish products, contains a 250-kilocurie plaque-form ^{60}Co source. The source is stored vertically under water but is used in the horizontal position,

*Marine Products Development Irradiator, Bureau of Commercial Fisheries Technological Laboratory, Gloucester, Mass. The authors acknowledge particularly the Gloucester Machine Shop for help in perfecting the design of the new bearings; John D. Kaylor, MPDI Supervisor for valuable ideas and effort; and Thomas J. Connors for Fig. 3.

†Built by the U. S. Atomic Energy Commission and operated by the U. S. Bureau of Commercial Fisheries; described in "Marine Products Development Irradiator" by P. Miller and J. Hubert, *Isotopes and Radiation Technology*, 1(4): 310–317 (1964); also in 3(4): 342–344 (1966).

being moved into the irradiation chamber by an elevator that rides on a 90° curved track (Fig. 1). The two roller chains that control the movement are guided by 20 sprockets — 10 for each chain with 9 above the water level and 1 below. The sprockets originally rolled on ball bearings, which failed and were replaced by solid bearings, which are much less likely to seize or fail suddenly than are ball bearings. Obviously, a failure of the bearings when the source is above water would create a dangerous radiation hazard. Wear in the new bearings is expected to be detected during scheduled inspections, so that repairs can be made before breakdown occurs.

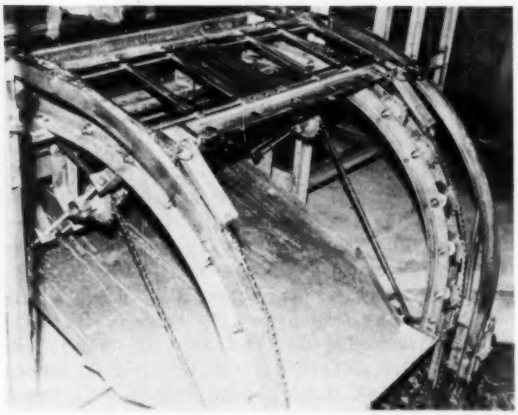

Fig. 1. Source-elevator mechanism showing model source holder in frame.

The technique used to replace the underwater bearings in the high radiation field generated by the ^{60}Co source and the bearing failures that made the replacement necessary are described here.

Underwater Replacement Technique

A completely underwater technique was developed and adopted for removing and replacing the bearings beneath the water level. The task proved simpler than anticipated and was considerably less costly than other procedures considered.

TOOLS

Since the pool is 16 ft deep, tool handles had to be about 18 ft long for convenient working. Many changes from one tool head to another are required for work of this type, and simple, standard connectors between handles and tool heads are therefore highly desirable. Also, the tools should transmit the "feel" of the work to the mechanic, who cannot see, for example, the nut he is unscrewing 18 ft away. Any unsteadiness of hand will be magnified by the 18-ft-long handle, so balanced tools in a vertical position, which tend, like a plumb bob, to assume the correct stance, are desirable. In keeping with this reasoning, most operations were performed with light tools mounted on flexible handles.

Some typical underwater tools are shown in Fig. 2. Stock aluminum conduit (*A*) was used for handles. It comes in 10-ft lengths with the ends already threaded fo. $1\frac{1}{2}$-in. National Pipe Thread (NPT) fittings. The three 18-ft lightweight handles made from it could be readily manipulated without floats.

A heavy handle was made of $1\frac{1}{2}$-in. stainless-steel pipe welded to a $\frac{3}{4}$-in.-square socket drive and used for loosening and tightening nuts.

An adapter (*B*) for fitting $\frac{1}{2}$-13 U. S. Standard (USS) threads to $\frac{1}{2}$-in. NPT threads was made by reaming and tapping a $1\frac{1}{2}$- by $\frac{1}{8}$-in. brass bushing; this operation is best done in a lathe so that the two sets of threads have a common axis.

A tool head (*C*) was designed for seating the chains on the sprockets after the brackets were bolted down during replacement. During assembly the chain loop hung loosely around the hub so that stiffness in the chain would not interfere with placing the brackets over the studs. Another tool head (*F*) was designed for starting the bracket hold-down nuts. It was made from a rubber stopper, which was bored and then

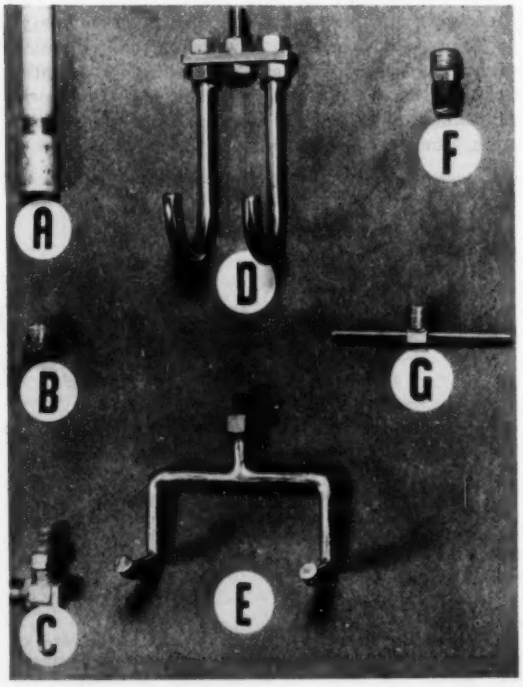

Fig. 2. Special tools used in replacing bearings at the bottom of the source storage pool.

fastened with a machine screw to a ½-in. brass pipe plug. When it was to be used, a nut was hand screwed on the stopper until it went about halfway through, the rubber conforming more or less to the shape of the threads. The tool head and nut were then lowered on an aluminum handle, and the nut was started on its stud. The rubber stopper served as a flexible coupling that allowed the nut to seek proper alignment for starting, even when the stud was not perfectly plumb.

The uses of tools *D*, *E*, and *G* are described below, in the Procedure.

Flat washers were picked up from the bottom of the pool and placed over the studs with a commercially available tool — the Radiarm Jr. Remote Handling Tong (Atomic Products Corp.).

As an underwater light we used a regular outdoor reflector spotlight in a rubber-sealed lamp base, attached to an extension cord. As a sight glass we used a floating box with a Lucite bottom.

PROCEDURE

The sprocket-bracket assemblies were easily removed as units after the three-plunger bumper for the source elevator had been hoisted out of place and the eight nuts holding the brackets unscrewed with a long-handled tool. Although stainless steel sometimes galls and seizes, no trouble was experienced with the nuts. A paper template showing the location of the boreholes was made so that the same dimensions could be maintained during reassembly.

The worn ball bearings were discarded, and the sprocket units were sent to a commercial machine shop for fabrication of the solid bearings (see below).

When the new bearings were received, the bolt-hole alignments were checked with the paper template. Two cap screws were fitted to the assemblies (Fig. 3) to support the sprocket axle on a paired hook (tool *D*, Fig. 2). Then, by hooking tool *E* over the two cap screws, we could control the assembly with sufficient accuracy to place it over the four hold-down studs. The chain had to be looped inside the bracket and the correct position maintained until washers were placed and at least one nut was started. The first assembly was placed without difficulty, but the cap screws interfered with starting the nuts. Therefore tool *G* was designed to slip into the holes from which the cap screws had been removed, and the brackets were placed without difficulty.

The nuts were started with tool *F*. The rubber stopper could then be pulled out without rotation and the nuts tightened with the socket wrench. Although the procedure for starting nuts had been considered a problem before work was started, only a few hours were required to install and bolt down the brackets.

OTHER METHODS CONSIDERED

Consideration was given to dismantling the sprocket by removing the nut from the axle with an electric impact wrench and driving the axle out with an electric hammer. A replacement axle bolt could be made, with the end machined down to the thread's root diameter, to serve as a mandrel for starting the nut under water. The electric tools would be protected with plastic film while under water. However, procurement of the necessary power tools and development of techniques for using them would have introduced a delay.

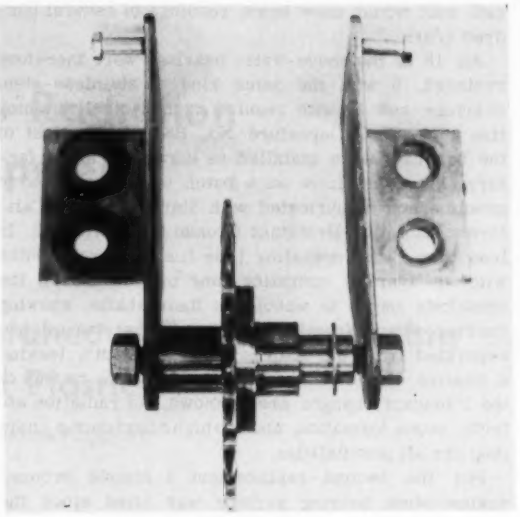

Fig. 3. Partially disassembled sprocket-bearing-bracket assembly before installation, showing cap screws.

The direct approach — loading the source into casks, removing them from the pool, and then pumping the water out of the pool — was considered as a last resort. It would have required a truck for bringing two 7-ton casks to the site with either two round trips for the truck or demurrage payment until the job was completed; a crane at a rate of $400 per day for lifting the casks; storage of the high-purity pool water; and incidental expenses of unloading the source material. Besides the costs, considerable time would have been lost.

Bearing Failures

The 20 original ball bearings were stainless steel (Barden No. RS 10SS) and had been degreased. Soon after the facility was completed, the bearings in one of the above-water sprockets froze — several others were found damaged — so that one chain ran off its sprockets and the source could not be moved. Fortunately this occurred when dummy sources were present; if an actual source should be stuck above water, fields of several thousand r/hr would be generated in the cell, and open tool holes through the

cell roof would show beam readings of several hundred r/hr.

All 18 of the above-water bearings were therefore replaced, 6 with the same kind of stainless-steel bearings and 12 with regular carbon-steel automotive type (New Departure No. RS 77R10). Most of the bearings were installed as lubricated at the factory, but a few from each batch were degreased in gasoline and relubricated with Standard Oil of California Radiation Resistant Grease No. NRRG 235. In less than 12 hr operating time (i.e., several months with an average operating time of 5 min/day), the sprockets began to wobble on their shafts, showing bearing wear. Examination showed that the oil had separated from the soap in both lubricants, leaving a waxlike deposit in the bearing race. The causes of the lubricant changes are unknown, but radiation effects, ozone formation, and too high a load on the chain loop are all possibilities.

For the second replacement a simple bronze-against-steel bearing surface was tried since the sprockets turn at only 14 rpm. Carbon-steel hubs, made to fit the sprockets and the supporting assembly, were bored to give a running fit on a standard Johnson No. 791GP bronze bushing. The bushings, which have a $^5/_8$-in. bore and are 1 in. OD by 3 in. long, were compressed between the angle-steel support frames of the sprockets; the original $^5/_8$-in. threaded shafts were used for bolts (Fig. 2). Since the original ball-bearing-mounting assemblies were interchangeable, the new hubs were made the same length. The hubs were provided with oil holes, which receive a drop or two of oil once a month.

The underwater bearings lasted about 2 years before excessive wobbling of the sprockets occurred, so apparently the water provided some lubrication. Since the steel-bronze bearings installed above the water level were operating well, similar ones were installed underwater. However, the hubs were made of stainless instead of carbon steel to avoid rusting. They were not prelubricated.

(MG)

Section

III

Isotopes and Radiation Technology

Process Radiation Development

Commercial Aspects of Instantaneous Radiation Curing of Reinforced Plastics*

By Walter Brennert and William F. Oliver‡

Abstract: Electron-beam irradiation has been used to cure glass-fiber-reinforced polyester laminates, prepregs, and molding compounds. The size and shape of parts that can be cured, commercially available machinery to provide sources of radiation, and processing costs are described here; physical properties of electron-beam and conventionally cured commercial laminates and prepregs are compared. With the advent of glass-fiber-reinforced polyester prepregs, radiation curing becomes a commercially feasible technique.

Electron-beam-radiation curing for the volume production of glass-fiber-reinforced polyester-plastic parts, which currently compose the overwhelming majority of all reinforced plastic composites manufactured, is stimulating a considerable amount of research and development of more economical methods for mass producing such parts with predetermined and reproducible high-performance qualities. Certain reinforced plastic parts already are, or shortly will be, produced in quantities above 100 thousand units per year, and plans calling for as many as 150 to 200 thousand parts per year are being made. At present about 30 thousand pieces per year per mold can be produced economically in a typical matched-die, hot-press molding operation.[1] With present equipment and materials, the time required to produce a single reinforced plastic molding is estimated to be between 100 and 260 sec — depending on the mechanical-press design features, available resins, and molding conditions, as well as on the thickness, size, and configuration of the molding.[2] Automated production operations and improved materials appear capable of reducing this time to as low as 32 to 59 sec per part, according to this study. While the shorter time range certainly represents a very substantial improvement in production capability, it is still a long way from matching that of metal-stamping operations, which have rates as high as 3.6 to 6 sec per part (600 to 1000 parts per hour) or faster.

Electron-beam-initiated curing of styrene-polyester resins has been described in several papers, with primary emphasis on coating applications.[3–5] Process advantages reported include essentially instantaneous curing in air§ at ambient temperatures for 100% polymerizable systems requiring no solvent removal, and, since heat is eliminated, the use of thermally sensitive substrates such as paper, wood, rubber, or plastics is possible. Boise Cascade Corp., under license from Ford Motor Co., is just starting up a pilot-plant facility, with a production unit scheduled to be operating next year[6] which will be capable of handling products up to 4 by 18 ft at rates as high as 60 linear ft/min. Boise Cascade is primarily interested in the coating of particle board and other wood products and will probably be using an acrylic system developed by Ford. Another company — Radiation Polymer Corp., Plainfield, Ill. — has announced the availability of radiation coating facilities for handling products up to 4 by 12 ft at speeds varying from 10 to 100 linear ft/min.

The possibilities of using electron-beam-initiated curing for production of glass-fiber-reinforced poly-

*This paper received the "Best Paper Award" in recognition of a contribution to the advancement of knowledge at the 22nd Annual Technical and Management Conference of the Reinforced Plastics Division of the Society of the Plastics Industry, Inc., Cleveland, Ohio, September 1966. The paper was published in the April 1967 issue (Vol. 23, No. 4) of *Society of Plastic Engineers Journal* and is reprinted here by permission. Editorial changes have been made for compliance to the format of *Isotopes and Radiation Technology*.

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‡Radiation Dynamics, Inc., Westbury, N. Y.

§All subsequent processing described in this paper is assumed to be at ambient temperature in air unless otherwise stated.

ester parts have recently been raised by Brenner, who reported some preliminary data at the September 1966 Cleveland meeting of the Society of Plastic Engineers (SPE). He summarized the current status of this work and provided most recent pertinent cost estimates for this novel production technology.

A considerable amount of experimental work has been done on the electron-beam-initiated curing of both glass-cloth and -mat wet lay-ups and prepreg* glass-fiber-reinforced polyester sheet stock. Various flat, corrugated, and tubular shapes have been irradiated, including filament-wound wet lay-ups and prepreg stock held in position on both thin metallic and nonmetallic mandrels. Experiments have also been conducted on the irradiation of chopped-glass-fiber-reinforced molding compounds that have been preformed into a number of simple shapes between two matching cold dies — roughly analogous to sheet-metal forming.

Cured laminates and chopped-fiber-reinforced structures have been produced almost instantaneously from uncured wet lay-ups and prepregs as well as from chopped-fiber-reinforced molding compounds by passing these materials, on a conveyor, under a 1.5-Mev Dynamitron† electron-beam accelerator (Fig. 1) to give an exposure of 3 to 10 Mrads. The thickness of these irradiated glass-fiber-reinforced shapes ranged from 10 mils to $\frac{1}{8}$ in. The accelerator was manufactured to specifications based on a production-rate capability equivalent to 9000 Mrads·lb/hr at 50% absorption efficiency and operation at 1.5 Mev. Its current range is 1.0 to 15.0 ma with a 12- to 24-in. beam scan of 100 cps.

The maximum thicknesses of unit-specific-gravity material which can be penetrated by the electron beam with this equipment and maintain equal ratios of entrance and exit doses are 90 mils for 0.75 Mev, 125 mils for 1.0 Mev, and 185 mils for 1.5 Mev. The specific gravities of various cloth and mat laminates of glass-fiber-reinforced polyester resin and of chopped-fiber-reinforced composites generally range from 1.4 to 2.0 or even higher, depending on the glass content and amount and types of fillers. The 1.5-Mev Dynamitron is therefore capable of producing radiation that can penetrate 0.132-in.-thick laminates of 1.4 specific gravity and 0.092-in.-thick laminates of 2.0 specific gravity.

The relations between the conveyor speeds with which a material can be moved under the electron beam (100 cps scan rate, 15 ma beam current) and the obtainable absorbed radiation doses, assuming 50% beam absorption efficiency and a specific gravity of 1, are 80, 40, and 16 ft/min for 1, 2, and 5 Mrads, respectively, for a 2-ft scan and half those rates for a 4-ft scan. The values for material with a specific gravity of 2.0 also receiving a 5-Mrad dose are identical, but the maximum penetration is half.

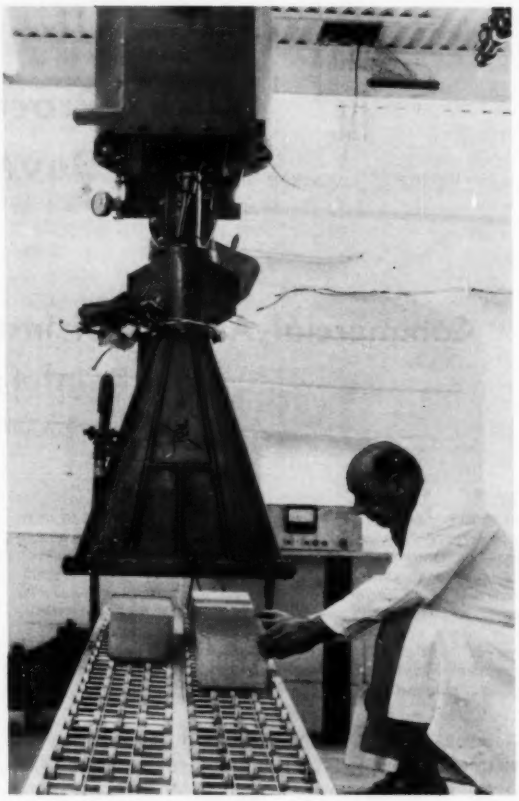

Fig. 1 Accelerator scan horn and conveyor table.

Larger sized (i.e., 3- and 4-Mev) Dynamitron electron-beam accelerators are available commercially with correspondingly increased service capabilities — e.g., a capacity for the 3-Mev Dynamitron of 14,000 Mrads·lb/hr at 50% absorption efficiency. The beam from this accelerator can penetrate a maximum of 370 mils of a unit-specific-gravity material or 185 mils of 2.0-specific-gravity material; other performance capabilities are doubled as well. The overwhelming majority of glass-fiber-reinforced plastic products now being manufactured have section thicknesses that can be effectively penetrated by a 3-Mev electron beam.

Electron-beam radiation-processing equipment is also commercially marketed by the High Voltage Engineering Corp., Burlington, Mass., which manufactures a range of particle-accelerator systems including relatively high-energy Van de Graaff machines and a recently developed low-energy insulating-core-transformer (ICT) line for industrial pro-

*Prepreg is a solid but uncured glass-reinforced plastic material.

†Manufactured and marketed by Radiation Dynamics, Inc., Westbury Industrial Park, Westbury, N. Y.

cessing. The General Electric Company, Milwaukee, Wis., produces resonant-beam transformers for radiation processing applications. Radiation Dynamics, Inc., has just introduced a small, relatively inexpensive electron irradiator called the "Dynacote," designed especially for production-line treatment of coated materials — metals, plastics, wood, paper, textiles, or ceramics. At the same time, the company has developed coating formulas that are particularly adaptable to the electron-beam curing technique.

Electron-Beam Curing of Wet-Lay-Up Glass-Resin Laminates

Experiments were carried out on the electron-beam-initiated curing of various wet-lay-up glass-cloth- and -mat-reinforced polyester-resin laminates. Uncured 24- by 12-in. lay-ups were placed on a conveyor and passed at rates varying from 9 to 18 ft/min under the 1.5-Mev Dynamitron accelerator beam. Operating conditions were arranged to give test samples radiation doses varying from 1 to 10 Mrads, and dose rates were varied from 1 to 5 Mrads per pass under the electron beam. The customary physical-strength measurements and chemical-resistance tests were made on these materials; ASTM methods were used whenever applicable. Basic physical-strength data on typical wet lay-ups cured by passage under a 1.5-Mev electron beam and the necessary radiation dosages required for curing are summarized in Table 1. The lay-ups were between 0.100 and 0.125 in. thick after curing. All but one were tack-free after the irradiation. The strength properties measured are comparable to those for similarly constructed laminates cured at elevated temperatures with peroxide catalysts. The actual electron-beam-radiation exposure times for these different laminate systems ranged from 2.5 to 6 sec depending on the conveyor speed. No subsequent heating (ovens) was used.

Pertinent chemical and electrical tests were also made on selected radiation-cured glass-fiber-reinforced polyester laminates. The initial results of one such series of chemical-resistance studies are shown in Table 2. Properties are similar to those published for peroxide-cured laminates. Some tests are still in progress.

Experiments have also been conducted to determine the elevated-temperature strength properties of radiation-cured polyester-resin laminates. Data are

Table 1 TYPICAL PROPERTIES OF ELECTRON-BEAM-CURED WET-LAY-UP GLASS-REINFORCED LAMINATES

Resin in cured laminate					
Type	Amount, wt.%	Reinforcement	Total dose for curing, Mrads	Room temp. flexural strength, psi	Room temp. flexural modulus, psi
Laminac 4128*	40	181 Volan A glass cloth	4–5	56,660	2.4×10^6
Laminac 4128*	50	2.5-oz. low-solubility-binder mat	4–5	20,800	1.7×10^6
Hetron 92†	45	181 Volan A glass cloth	3–4	54,730	1.6×10^6
Laminac 4104*	40	181-136 glass cloth	3–4	61,500	2.8×10^6
Glidpol 1001‡	42	181-136 glass cloth	4–5	58,080	2.8×10^6
AP 285§	40	181 Volan A glass cloth	6–8	70,420	3.3×10^6
PE 327¶ with aluminum hydrate C331 filler	40 30	1.5-oz. low-solubility-binder mat	4–5	26,200	1.4×10^6

*American Cyanamid Company, Wallingford, Conn.
†Hooker Chemical Corp., North Tonawanda, N. Y.
‡The Glidden Company, Cleveland, Ohio.
§Rohm & Haas Co., Philadelphia, Pa.
¶Allied Chemical Corporation, Morristown, N. J.

Table 2 TYPICAL PROPERTIES OF RADIATION-CURED POLYESTER GLASS-MAT-REINFORCED LAMINATES*

(0.012-in.-Thick Test Specimens; 28 wt.% T219 Glass Mat)

	Flexural strength, psi						Weight gain, %	
Means of curing	Initial at room temp.	2 weeks in H_2O, 212°F	4 weeks in H_2O, 212°F	4 weeks in concentrated HCl, 212°F	4 weeks in 50% H_2SO_4, 160°F	4 weeks in saturated alum solution, 212°F	4 weeks in H_2O, 212°F	4 weeks in concentrated HCl, 212°F
Irradiation, room temp., 5-Mrad dose	22,600	7,100	7,300	5,600	15,200	6,000	+0.8	−18.9
Benzoyl peroxide, 0.75%, 15 min at 266°F†	23,600	6,700	7,400	5,300	14,500	6,300	−0.7	−20.6

*Laminac 4119, American Cyanamid Company.
†Cyanamid technical literature.

presented in Table 3 for Laminac 4104 glass-fiber-mat-reinforced laminates at temperatures up to 250°F. A comparison with similar conventionally prepared laminates shows similar physical-strength properties at elevated temperatures.

Table 3 ELEVATED-TEMPERATURE PHYSICAL-STRENGTH PROPERTIES OF RADIATION-CURED LAMINAC 4104 GLASS-MAT-REINFORCED LAMINATES

(Three Plies, 1.5-oz. Low-Solubility-Binder Mat; 0.120-in.-Thick Test Specimens)

	Flexural property, psi					
	Room temp.		176°F		250°F	
Means of curing	Strength	Modulus	Strength	Modulus	Strength	Modulus
Irradiation, room temp., 5-Mrad dose	4.25×10^4	1.59×10^6	3.26×10^4	1.35×10^6	2.21×10^4	0.88×10^6
Benzoyl peroxide, 0.5%, 10 min at 235°F	4.54×10^4	1.78×10^6	3.49×10^4	1.43×10^6	2.07×10^4	0.94×10^6

These tests show that electron-beam curing of glass-fiber-reinforced wet-lay-up laminates results in composites with physical-strength properties and chemical characteristics equivalent to those of peroxide-initiated heat-cured laminates. The exceptionally short cure cycles offer most interesting commercial possibilities.

Electron-Beam Curing of Commercial Prepregs

Experiments were carried out on the electron-beam-initiated curing of two types of commercially marketed glass-fiber-reinforced polyester prepreg materials: Uniroyal Chemical Division's Vibrin-Mat Type G 1600 and Standard Oil of Ohio Company's Structoform. Uncured 24- by 12-in. sheets were passed under a 1.5-Mev electron-beam accelerator at rates varying from 9 to 18 ft/min. The radiation doses were varied from 1 to 10 Mrads. The Vibrin-Mat G 1600 test samples as supplied usually contained catalyst, were 0.080 in. thick, and weighed approximately 300 g/sq ft. They had been cold pressed at 200 psi before irradiation to remove wrinkles. The Structoform prepregs were obtained without catalyst and had been pressed at 300°F and 400 psi by the manufacturer. Radiation dose rates were varied from 1 to 5 Mrads per pass. For doses greater than 5 Mrads, two passes were used, one a 5-Mrad pass and one at the level required to make up the indicated total.

Total radiation-exposure times ranged from 2.5 to 6 sec per pass depending on the conveyor speed. The irradiated materials were tested for physical strength and chemical resistance by applicable ASTM methods.

Table 4 indicates that doses of 5 to 6 Mrads are capable of curing Vibrin-Mat G 1600 and Structoform prepreg materials. Very short curing cycles, of the order of seconds, can be used because curing can be accomplished merely by passing the materials under the electron beam at rates sufficient to give the necessary radiation doses. The physical-strength properties of the products approach those of materials processed by conventional hot-press, matched-die molding. This is somewhat surprising for Vibrin-Mat prepreg laminates, which contain only 24 wt.% glass fibers compared with the 30 wt.% reported by the producer. Water-absorption, chemical-resistance, and electrical-insulating tests on electron-beam-cured glass-fiber-reinforced polyester prepregs indicate that the shelf-life and storage limitations of conventionally compounded materials are eliminated,

Table 4 PROPERTIES OF RADIATION-CURED PREPREG MATERIALS

(Ambient Temperature in Air)

Material	Dose, Mrads	Flexural modulus, psi	Flexural strength, psi	Tensile strength, psi	Ultimate elongation, %
Vibrin-Mat G 1600*	1	560,000	11,000	8,360	3.2
	2	970,000	19,020	8,530	2.4
	3	1,250,000	21,940	10,530	2.7
	4	1,170,000	21,910	9,490	2.8
	5	1,160,000	22,720	10,830	3.1
	6	1,260,000	22,070	9,430	2.3
	7	1,180,000	20,500	10,030	2.7
	8	1,280,000	24,140	10,630	3.0
	9	1,130,000	20,230	10,140	2.8
	0†	1,500,000	26,000	15,000	-
	0‡	1,307,240	23,560	12,060	2.7
Structoform§	1	1,260,000	22,590	12,480	3.7
	4	1,689,000	33,550	15,670	4.1
	5	1,520,000	29,880	14,060	3.8
	6	1,720,000	34,170	13,920	3.7
	8	1,680,000	34,220	15,260	4.1
	10	1,390,000	28,570	15,680	4.2
	0¶	1,800,000	30,000	15,000	-

*Contained catalyst as supplied commercially; glass content 24 wt.%.

†Manufacturer's data for hot-press-molded G 1600 prepreg mat laminates; glass content 30 wt.%.

‡Experimentally obtained data for hot-press-molded G 1600 prepreg mat laminates; glass content 24 wt.%.

§Structoform prepregs tested did not contain catalyst; glass content 33 wt.%.

¶Manufacturer's data for hot-press-cured Structoform prepreg laminates; glass content 33 wt.%.

resulting in various obvious benefits to both the prepreg manufacturer and the processor.

The data of Table 4 also suggest that the incorporation of peroxide catalysts (which are not required for electron-beam curing) in such prepreg materials does not affect the electron-beam-initiated curing of such materials. This has been confirmed by considerable additional testing.

From the prepreg manufacturer's point of view, the exclusion of catalysts permits much greater latitude in manufacturing conditions and virtually eliminates problems of premature curing during manufacturing as well as loss of material by progressive deterioration, which occurs during prolonged or improper storage conditions and which impairs molding characteristics.

Electron-Beam Curing of Commercial Molding Compounds

A limited amount of experimentation has been devoted to the study of radiation curing of glass-fiber-reinforced polyester molding compounds. A typical compound, which contains a catalyst and is glass fiber reinforced and mineral filled, is an alkyd molding putty, Plaskon 760, commercially available from Allied Chemical Corporation. This material was shaped into flat sheet stock by cold pressing between flat platens with stops to produce 0.060- and 0.090-in.-thick sheets. These sheets were placed on a conveyor and irradiated with the electron beam, as before. Strength properties were very similar to those of conventionally hot-press-molded materials (Table 5).

Table 5 PROPERTIES OF RADIATION-CURED PLASKON 760 ALKYD MOLDING COMPOUND

(0.090-in.-Thick Flat Sheet Test Specimen)

Physical property	Radiation-cured, room temp., 5-Mrad dose	Hot-press-molded, 300°F, 1000 psi
Flexural strength, psi	14,700	15,100
Flexural modulus, psi	2.0×10^6	2.2×10^6
Tensile strength, psi	6,800	7,100
Compressive strength, psi	24,300	25,000
Impact strength, Isod, ft-lb/in., notch	3.8	3.8
Dielectric constant, 1 Mc	5.8	5.9
Dissipation factor, 1 Mc	0.009	0.011
Barcol hardness	65	69

Costs and Application Potentials

The average installed price quoted for a 1.5-Mev Dynamitron electron-beam accelerator is $180 thousand exclusive of shielding, and recent experience indicates the cost of a complete, properly shielded, facility to be about $230 thousand. Assuming a 7-year amortization ($16.50 per hour) and accelerator operation of 2000 hr per year, operating costs are estimated to be $34 per hour. Other assumed costs are utilities, including electricity and water, at $1.50 per hour; an operator at $3.00 per hour plus 100% overhead; maintenance $7.00 per hour; and 10% for miscellaneous expenses. For 4000 hr of operation per year, the operating cost would be about $26 per hour.

Using $34 per hour for the accelerator operating cost and assuming 50% utilization and a 5-Mrad dose, we find the cost of electron-beam-initiated curing to be 1.89 cents per pound, or 0.53 lb of product per penny. The total production capacity will be 1800 lb/hr for a 5-Mrad dose. The present cost of benzoyl peroxide paste (50% active) in large quantities is quoted at $0.95 per pound. The catalyst cost for conventional curing amounts, therefore, to between 0.95 and 1.90 cents per pound of product, depending on the amount of catalyst used; this does not include operator and heating costs. Thus it can be readily seen that electron-beam-radiation curing is a processing technique essentially competitive with conventional-catalyst methods provided that production quantities are large enough to warrant the capital investment involved and that product geometries are suitable for operation of this process.

The processor of reinforced plastics now has available a uniquely effective processing tool, particularly suited to large-volume, high-production-rate molding operations. This tool substantially exceeds, in both production capacity and rate, currently used conventional processing equipment for thermosetting plastics. It is most promising, of course, for large-scale production of parts suited for continuous processing operations, e.g., pipe and sheet stock, and is amenable to complete automation.

The processor is also freed from material shelf-life and storage problems and can therefore manufacture a most uniform quality product, since prepreg molding qualities are quite independent of storage time. Automation, coupled with much higher production rates, offers most intriguing cost-reduction possibilities and should result in the enhancement of the competitive position of thermosetting laminates, which have characteristics superior to those of the more widely used thermoplastics. In the electron-beam processing of the otherwise "dirty" and wasteful wet lay-ups, as well as of prepreg and molding compounds, irradiation cost will be offset by attainment of much higher production speeds; elimination of catalyst, ovens, and other equipment; and reductions in labor costs. Process advantages increase in direct proportion to the number of parts processed.

The main limitations of radiation curing of wet lay-up and prepreg materials relate to applicable product shapes. At present, this processing technique appears to be most useful for relatively simple geometric shapes such as rods, tubes, and cylinders. It appears to be a natural for the manufacture of both

plain and corrugated sheeting, pipes, and other shapes, including containers, ducting, body-shell parts, and housings. Filament-wound reinforced plastic parts have a geometry that is well suited for electron-beam-radiation processing, subject to the size of present accelerator equipment.

The electron-beam irradiation of more complicated geometric shapes, e.g., undercuts and reentrant curves, requires more sophisticated design of mechanical equipment. Reflectors may be required to direct electron beams into certain areas, and filters can be developed to screen the radiation from others. Rotation of certain geometrically simple product shapes, such as pipes, during electron-beam irradiation has been accomplished quite readily with suitable mechanical equipment and should be feasible for more complicated shapes with proper mechanical engineering. Greater flexibility in the design of the head of the electron-beam accelerator, so as to permit its movement during irradiation, should further enhance the range of shapes that can be satisfactorily irradiated. These problems, which are essentially engineering, are capable of solution in one way or another, given the impetus of more economic, high-speed, automated large-volume-production possibilities.

Economic feasibility is dependent on the anticipated production volumes, and both present and anticipated production volumes must be large enough to justify the considerable capital outlay required for the purchase and installation of an electron-beam-irradiation facility. Given such economic justification, the application of electron-beam irradiation for the curing of wet-lay-up glass and prepreg fiber laminates and molding compounds is considered to offer unique opportunities to alert processors, not only for the development of volume markets for novel products, but also for the more efficient and economic manufacture of current volume product lines. (REG)

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Chlorination of Toluene Using an Internal Ionizing-Radiation Source*

By Carol H. Collins†

Abstract: Toluene can be chlorinated either by side-chain substitution or ring addition. Ionizing-radiation-initiated and photoinitiated chlorination reactions yield the same products under the same reaction conditions. Ring addition predominates at temperatures below 20°C, while side-chain substitution occurs essentially exclusively at temperatures above 80°C, producing benzotrichloride. Thus the choice of initiation source to be used for the side-chain chlorination of toluene would be determined by the most efficient way to optimize the chosen reaction conditions and by the comparative economics that would be obtained. No valid comparisons of the type of source can be made for the reaction of chlorine and toluene, as sufficient information is not available in the literature. However, comparisons of energy utilization for the chlorination of benzene indicate that gamma radiation is 8.5 times as effective as the most efficient ultraviolet light source. Ionizing radiation would compare even more favorably with less efficient light sources. That this is true for the chlorination of toluene would have to be proved, however, since yields (G values) for the chlorination of both benzene and toluene depend on the reaction conditions as well as on the radiation source intensity. In the determination of the preferable type of source, consideration of linear energy transfer (LET) effects indicate that a radiation source of low LET would produce fewer unfavorable side effects (such as radiolysis of benzene or toluene) while maximizing the yield of chlorination products. Although either a beta- or a gamma-radiation source could meet the requirement of low LET, a properly constituted low-energy internal beta source would offer the potential of more efficient energy utilization coupled with a favorable minimization of radiation shielding requirements.

This article summarizes currently available information about chlorination reactions, with particular emphasis on the chlorination of toluene, and dis-

*Based on USAEC Report NYO-3718-1, Development of a Radioactive Pebble Bed Chemical Reactor System. Phase I, by C. H. Collins, H. T. Cullinan, C. C. Thomas, Jr., and S. Weller, Sept. 15, 1966.

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cusses the possibility that an economically competitive process can be developed for the production of benzotrichloride (a product of significant commercial attractiveness) using an internal beta-emitting radiation source.

The use of radioisotopic energy sources as initiators for chemical reactions of commercial significance is of interest to both the Atomic Energy Commission and the chemical industry. To be considered commercially attractive, the ionizing-radiation-initiated reaction must be a process which (1) results in a product not obtainable by another method; (2) can be carried out more conveniently with ionizing radiation; or (3) is more economical when carried out with ionizing radiation. Although there are reactions initiated by ionizing radiation which produce a product not obtainable by other means, in most cases radiation initiation must successfully demonstrate its worth in direct comparison with other means, e.g., photolytic, thermal, or thermocatalytic processes. These radiation processes must therefore show high G values,* i.e., they must be exothermic, chain processes. Chlorination reactions appear to satisfy these requirements.

Radiation Chlorination of Aromatics

In a general sense, an aromatic hydrocarbon may react with chlorine in four ways:

1. Substitution of chlorine for a hydrogen attached to the ring
2. Replacement by chlorine of a substituent, other than hydrogen, on the ring
3. Substitution of chlorine for a hydrogen from an alkyl substituent group attached to the ring
4. Addition of chlorine to the ring, resulting in a loss of aromaticity

The first process occurs (whether desired or not) in the presence of Friedel–Crafts catalysts, such as ferric chloride or aluminum chloride, or in the presence of iron or other metals that can react with chlorine to form chlorides.[1]

The second process occurs readily, replacing certain substituent groups, e.g., bromo-, on the aromatic ring and less readily with other substituent groups, e.g., di- or trichloromethyl- (Ref. 2). Such substitution can result in the loss of a desired substituent. Although this reaction often occurs during light- or radiation-promoted reactions, it is an ionic reaction.

It is well established that the third and fourth processes proceed by free-radical mechanisms.[3] Thus these processes can be initiated by typical free-radical means, including the use of peroxides, heat, or radiation (either "low energy" radiation such as ultraviolet light or "high energy" radiation such as gamma rays), and are inhibited by free-radical inhibitors such as oxygen. The reactions are also reported to occur in the dark, but to a markedly decreased extent. However, an addition reaction has been reported to occur in the absence of light in the presence of catalysts such as dilute sodium hydroxide.[4]

In most cases the use of the different forms of radiation, e.g., ultraviolet light or gamma radiation, to induce a free-radical chain reaction yields the same mixture of products, provided the conditions of the reaction are the same in both cases. This observation, reported by Lind and coworkers[5] as early as 1926, has been verified for a wide variety of reactant mixtures.[3] It can be explained if the excited species produced by ionizing radiation are assumed to be essentially identical to those produced by ultraviolet excitation.[6] The ions formed by the ionizing radiation would thus not enter directly into chemical reactions but would first be neutralized to give an excited molecule which then behaves similarly to the excited molecules produced by the photochemical process.

With benzene and alkylaromatic compounds, the composition of the reaction product mixture and, for the alkylaromatics, the relative amounts of side-chain substitution and ring addition that occur in the free-radical reaction are reported to be dependent on several factors—temperature (higher temperatures favor side-chain substitution of alkylaromatics), chlorine concentration (higher chlorine concentrations enhance the addition reaction), and the intensity of the radiation source.

ADDITION OF CHLORINE TO BENZENE

The photoinitiated addition of chlorine to benzene was first reported in 1825 by Faraday,[7] and the "optimum conditions" for the reaction were discussed by Mitscherlich[8] in 1835. Several kinetic studies on the reaction velocity[9] had already been performed when van der Linden[10] reported that several isomeric forms were present among the reaction products. The potential use of the addition products in an insecticide was first cited[11] in a patent in 1935, but widespread use in insecticide preparations had to await the discovery, in 1945, that the γ isomer of hexachlorocyclohexane (γ-benzene hexachloride, popularly known as Lindane or Gammexane) and only the γ isomer was biologically active.[12]

Since 1945 a great number of patents† have been issued dealing with potential modifications (light intensity, optimum wavelength, effects of additives, effects of solvents, type of solvent, effect of temperature, length of reaction, concentration of chlo-

*The G value is a measure of the number of molecules reacted or produced per 100 ev of radiation energy absorbed.

†Any list of patents on this or subsequent topics would be too long and, undoubtedly, incomplete. Thus no attempt will be made to enumerate patents other than those of significant interest to the discussion.

rine, and the rate of chlorine addition) of the photochlorination reaction with benzene. In addition, numerous papers in the literature deal with these effects on the addition reaction. Unfortunately, many of the papers and patents indicate conflicting results, although nearly all are concerned with increasing the content of the γ isomer (γ-hexachlorocyclohexane) while maintaining a high overall yield.

Average yields in the photochemical reaction are 13 to 15% γ isomer with an overall yield near 100%. Higher γ-isomer yields (20% or more) have been reported, but these occur generally at the expense of a poorer total or overall yield. The highest γ-isomer yield reported to date is 42% at 100% conversion.[13] However, this result could not be reproduced by other workers.[14,15]

Perhaps the most thorough recent study of the photochlorination of benzene was carried out by Roberts and coworkers[14,15] in their comparison of the ultraviolet- and radiation-initiated addition reaction of chlorine and benzene. Using a mercury lamp with maximum emission at 3650 A (the most efficient wavelength for chlorine dissociation is at 3350 A), these workers observed a quantum yield* for disappearance of chlorine of 1580 with no additives or pretreatment. The quantum yield rose to a maximum of 8670 when purified benzene had been purged and presaturated with chlorine for 1 hr. Other workers[16] have reported quantum yields of 2500 under their optimum conditions. Since the British workers[14,15] were using "optimum" light conditions, they could not increase either the overall yield or the percentage of γ isomer present by increasing the reaction time or by the use of additives, although both these methods had been cited as effective by others.[13,17] However, Fürst and coworkers,[17] as well as many others, carried out their experiments with a less efficient light source, so the effect of additives, especially those of the photosensitizer type, may be real with these less efficient initiation conditions.

The first reports of the use of ionizing radiation to initiate the addition of chlorine to benzene using X rays were[18] in 1924, and using radon alpha radiation[19] in 1930. The potential availability of large radiation sources coupled with the potential commercial demand for γ-hexachlorocyclohexane as an insecticide motivated several studies of the gamma-radiation-induced chlorination of benzene in the early 1950's.

In the United States, studies at the Fission Products Laboratory at the University of Michigan[20,21] indicated that the chlorination of benzene using a 300-curie ^{60}Co source was so vigorous that considerable dilution of the benzene with an "inert" solvent — carbon tetrachloride — was necessary. Added solvent, also present in most photochlorination reactions, helps to prevent the deposition of solid products and to control the heat of reaction. Yields (G values), expressed as molecules of chlorine reacted per 100 ev of radiation absorbed, ranged from 19,000 to 114,000 with a G(-Cl_2) = 85,000 reported[20,21] for the "standard" conditions when chlorine is bubbled through a 20% solution of benzene in carbon tetrachloride in the presence of a ^{60}Co radiation source (dose rate 6.16×10^{16} ev/(g)(min). Under these conditions the percentage yield of hexachlorocyclohexane, based on benzene, is 97%, with a γ-isomer content of 11 to 13%.

A more recent study at the University of Minnesota,[22] using undiluted benzene in a stirred-tank multipass (continuous) reactor system with a 10,000-curie ^{60}Co radiation source, reported G values of 21,800 with a dose rate of 3.06×10^{17} ev/(g)(min) and 61,300 with a dose rate of 6.40×10^{16} ev/(g)(min). The γ-isomer content of the reaction product is reported to be 19 to 22% from an infrared spectroscopy determination. These authors propose that the addition reaction proceeds by a free-radical chain-reaction mechanism — first order with respect to chlorine concentration and independent of both temperature in the range investigated (15 to 30°C) and surface-to-volume ratio of the reaction vessel (varied by a factor of 2.2). The reaction rate was reported to be dose-rate dependent, proportional to $I^{0.2}$ rather than showing the commonly accepted square-root dependence.

Research on the addition chlorination of benzene at the Wantage Laboratories of the Atomic Energy Agency in Great Britian[15,23] with a 700-curie ^{60}Co source also indicated an inverse dependence of the G value on the dose rate (cited to be a square-root dependence). These authors reported G values for the disappearance of chlorine in its reaction with pure benzene to be 560,000 at 4.1×10^{15} ev/(g)(min), 370,000 at 5.6×10^{15} ev/(g)(min), and 180,000 at 8.9×10^{16} ev/(g)(min). These yields are considerably higher than those of other researchers.[20-22] The G values were reported[15,23] to depend on the benzene concentration (in carbon tetrachloride) but to be independent of reaction temperature (−72 to +80°C). A slight effect on the γ-isomer content of the product (increasing from 16 to 18%) was reported for the lowest temperature used (−72°C). Otherwise, the γ-isomer content ($15.3 \pm 1.6\%$) was unaffected by dilution, dose-rate changes, or changes in the reaction time.

These results were compared with those obtained using ultraviolet radiation.[14,15] Converting the observed quantum yield (8670 at 3650 A) to a radiation yield (the quantum energy at 3650 A is approximately 3.40 ev) gives a G value of 42,500 for the photochemical reaction. Since all other aspects of the process are essentially identical (gross yield, percentage γ-isomer content), these workers concluded that gamma initiation for the same energy absorption was approximately 8.5 times as effective

*The quantum yield is a measure of the number of molecules reacted or produced per quantum of light energy absorbed.

as photochemical initiation (when the illumination is in the most favorable wavelengths). It should be emphasized that this factor of 8.5 may be an artifact due to the reliance of these workers on the inverse-square-root law, since subsequent investigation[22] has indicated a 0.2 power dependence on intensity. On the other hand, this factor may reflect the effect of additional types of initiation steps due to the excitation of both benzene and chlorine with gamma radiation as compared to excitation of the chlorine alone when using photoinitiation.

Extensive studies in the USSR have also compared ultraviolet initiation with gamma initiation from ^{60}Co for the reaction of chlorine with benzene. Early reports[24] indicated identical γ-isomer contents, independent of temperature. Later investigations[25] reported higher γ-isomer contents (up to 30%) using ionizing radiation of several types (gamma from ^{60}Co, mixed gamma and beta radiation from a fission-fragment source, and beta from a ^{32}P source). The G values were reported as 210,000 for a ^{60}Co gamma source with a dose rate of 8.9×10^{15} ev/(g)(min), 380,000 for a ^{32}P beta source with a dose rate of 1.8×10^{15} ev/(g)(min), and 46,000 for a fission-fragment source with a dose rate of 1.1×10^{17} ev/(g)(min). These workers suggested that higher α- and γ-isomer contents (at the expense of the β and δ isomers) from the radiation reaction would make radiation initiation more favorable than photochemical initiation.

The potential utility of the radiation-initiated addition of chlorine to benzene was such that several analyses of the process economics were published[26,27] during the 1950's and several patents issued.[28,29] However, recent declines in the market potential of γ-hexachlorocyclohexane have tended to discourage further commercial consideration of the radiation-initiated process.

REACTION OF CHLORINE WITH TOLUENE

Addition of chlorine to the toluene ring structure under the influence of sunlight to produce a compound described as $C_7H_8Cl_8$ was reported[30] in 1867, while preparation of benzyl chloride from toluene and chlorine using sunlight was reported[31] in 1885. However, these and the many subsequent early papers disagreed not only about the relative amounts of addition to the ring vs. substitution on the side chain to be observed in these free-radical reactions, but also about the conditions under which either or both of these reactions could be observed. Thus Book and Eggert[32] reported that side-chain substitution is facilitated at −80°C, while Bergel[33] reported that substitution is favored at elevated temperatures. The addition reaction is also reported to be the preferred mode at −15°C (Ref. 11) and at 110°C (Ref. 34). However, most early studies of the chlorination of toluene[35] were carried out at room temperature where a mixture of ring-addition and side-chain-substitution products was observed. Recent and more exacting work on the photochlorination reaction has indicated that the ring-addition reaction occurs preferentially, if not exclusively, at low temperatures[36,37] (−40 to +20°C) and that side-chain substitution is the exclusive reaction at high temperatures[2,38–42] (>80°C) as well as in the vapor phase,[43] while at ambient temperatures (30 to 60°C) mixtures of ring-addition and side-chain-substitution products are obtained.[42,44,45] The ratio of ring-addition to side-chain substitution observed in these mixtures appears to depend quite strongly on the concentration of chlorine reactant and on the degree of side-chain chlorination already present. With equal amounts of chlorine and toluene, 20% ring-addition products are reported[42] at 40°C. Higher yields of ring-addition products (greater than 80%) are reported at 15°C for a 2:1 toluene-to-chlorine ratio.[45] Yields in the low-temperature ring-addition reaction are reported to be 80% or higher,[37] while high-temperature liquid-phase side-chain-substitution reaction yields are also reported[42] to be at least 80%. In the vapor phase the only product is benzyl chloride, reported to be obtained in high yield.[43]

The rate of the photochemical side-chain-substitution reaction has been reported to be first and half order with respect to toluene and chlorine concentrations, respectively, and to depend on the square root of the intensity.[40,42] The side-chain-substitution reaction proceeds to nearly complete conversion to benzyl chloride before benzal chloride appears. Complete chlorination eventually yields benzotrichloride. Increases in the reaction temperature facilitate the production of benzal chloride and benzotrichloride. The relative rates of the photochemical side-chain-chlorination reaction on toluene, benzyl chloride, and benzal chloride, have been reported to be 1.0:0.09:0.005 in a static reaction system[46] and 1.0:0.17:0.029 in a continuous reactor system,[42] both at 100°C. The rate of substitution of chlorine on the methyl groups of toluene and on the chloromethyl group of benzyl chloride increases by a factor of 2 at lower temperatures (40°C), but the rate of chlorine atom reaction with benzal chloride is reported to be unaffected.[42] Roberts and coworkers,[14,15] in their studies of the photochlorination of benzene, reported the highest yields with photolytic radiation of wavelength nearest to that of the chlorine bond dissociation[42] (3350 A).

The first report of the use of gamma radiation to initiate the chlorination of toluene came from the Fission Products Laboratory at the University of Michigan.[20,21] In reactions carried out at 20°C, both ring-addition and side-chain-substitution products were observed. The G value for these reactions, reported as the amount of chlorine consumed, was 17,000. The side-chain-substitution reaction initiated by gamma radiation was found to depend on the square root of the chlorine concentration as well

as on the square root of the source intensity, dependences similar to those reported for the photochemical substitution reaction.[40,42] The ring-addition reaction was reported[20,21] to be second order in chlorine concentration and did not directly follow the square-root law for dose-rate dependence.* In distinction from the photochemical reaction,[42] both the addition and substitution reactions were reported to show an increased rate with increasing temperature.[20,21]

Although several workers had reported the occurrence of mixed ring-addition and side-chain-substitution products when the photochlorination reaction was carried out at ambient temperature,[35,44] the University of Michigan researchers, by assuming an extrapolation of high-temperature photochlorination results (only side-chain chlorination) to their ambient-temperature reaction conditions, concluded that the product mixture obtained with gamma radiation (mixed side-chain-substitution and ring-addition products) was different from that obtained in the photolytic reaction. It was suggested[26] that this difference in reaction product mixture would open up the field of radiation synthesis since the different products meant different reaction mechanisms. This most interesting but, unfortunately, premature concept was quickly contradicted. Cox and Swallow[47] as well as Rosen[45] compared the products from the gamma-radiation-initiated and the ultraviolet-light-initiated reaction of gaseous chlorine bubbled into liquid toluene at 15°C. Identical product spectrums for the two types of initiation were reported[45,47] when reaction conditions were identical. Rosen carried his reaction to a high consumption of chlorine. He reported the major product from both modes of initiation at 15°C to be methylhexachlorocyclohexane, with minor amounts of benzyl chloride. Cox and Swallow,[47] using different reaction conditions and a low conversion, reported identical amounts of benzyl chloride as the major product, with ring-addition products being obtained in relatively minor amounts whether the initiation resulted from ultraviolet, ^{60}Co gamma, or fast-electron (from a 2-Mev Van de Graaff generator) irradiation. These workers reported the G value for benzyl chloride production with gamma radiation to be about 10,000. Under other conditions the G value, measured as chlorine consumed, ranged[48] from 10,000 to 100,000. The rate of the side-chain-substitution reaction was reported[47] to have a square-root dependence on both the chlorine concentration and the radiation intensity. The addition reaction rate was reported[48] to be second order with respect to chlorine concentration but to depend on the square root of the dose rate. In contrast to reports by Harmer and coworkers[20,21] of a severe inhibition of the gamma-radiation-initiated chlorination of toluene by benzyl chloride, Cox and Swallow[47] reported that the chlorination reaction could be carried out using benzyl chloride as solvent without depressing the reaction rate. They also reported that the chlorination of benzyl chloride to produce benzal chloride occurred at a rate approximately 10% of that of the rate of chlorination of toluene. This value is very close to those reported for the photochlorination reaction.[42,46] These results of Rosen[45] and of Cox and Swallow[47] were expanded by some Japanese workers,[49] who have confirmed the identity of product spectrum between radiation initiation and ultraviolet-light initiation over a wide range of temperatures. They also report that at 80°C and above, with either type of initiation, the side-chain-substitution reactions are the only ones observed, while at room temperature the ring-additon reaction predominates although some side-chain substitution still occurs.

Both the ring-addition and the side-chain-substitution products from the chlorination of toluene have commercial importance. However, no recent work has been carried out in this country on the application of radiation to their production.

REACTION OF CHLORINE WITH OTHER ALKYLAROMATICS

The xylenes, ethylbenzene, cumene, mesitylene, isopropylbenzene and diisopropylbenzene have all been successfully photochlorinated (Refs. 2, 37, and 50 to 53). Most of these reactions have been carried out at elevated temperatures so that side-chain substitution is the major, if not exclusive, reaction. Good yields have been reported although there is a decrease in the rates of successive substitutions,[51,53] similar to the rate decreases observed with toluene and its α-chlorosubstituted products. As with toluene, when the photochlorination reaction is carried out at low temperature, ring-addition reactions are observed.

Partial photochlorination of multialkyl-substituted aromatics reveals that, for the most part, monosubstitution will occur on all substituents before the reaction which gives disubstitution on the same substituent begins.[2] In a similar manner the side-chain-substitution reaction proceeds almost completely to the multi-disubstituted product before appreciable amounts of trichloro product appear. This observation has been useful in the development of photo-initiated reaction conditions to produce partially chlorinated alkylaromatic products.[54]

The presence of chlorine both in the group being acted upon and in the group para to it lowers the reactivity of the C–H bond being acted upon.[2,50] With certain alkylaromatics, the product with fully chlorinated side chains cannot be obtained or is observed only in very small yield along with other products, often those resulting from ring substitution.[2,50] The

*Harmer's thesis[21] indicates a dependence in the addition reaction of $I^{0.6}$, while papers from the Fission Products Laboratory[20] report a first-power dependence on intensity. No explanation is given for this discrepancy.

presence of other substituent groups on the ring can also cause nuclear substitution reactions or scission, in which the chlorosubstituted methyl group is replaced.[2,50]

Very little research has been reported on the radiation-initiated chlorination of alkylaromatics other than toluene. The research to date on this topic has been carried out at the Fission Products Laboratory at the University of Michigan.[26,55] Mesitylene, ethylbenzene, *m*-xylene, and several methylnaphthalenes were treated with chlorine while being exposed to a 3000-curie ^{60}Co source. Distillation analysis showed that, at the temperature used, 15 to 20°C, the products were mainly those resulting from ring-addition reactions.[55]

Although no supporting data are available, it may be safely assumed, by analogy with the photochlorination of alkylaromatics and from the near identity of products from both modes of initiation when the substrate is toluene, that a change in distribution from ring-addition products to side-chain-substitution products will occur as the reaction temperature of the ionizing-radiation-initiated chlorination reaction is increased to the levels used to obtain side-chain substitution in the photochlorination reaction.

REACTION MECHANISMS

The above reviews of the chlorination of benzene, toluene, and other alkylaromatics have shown that no special effects, such as differences in the product distribution, can be attributed to the mode of initiation of the reaction. Thus, for the chlorination of toluene, the product ratios and even the product yields have been shown to be essentially identical, independent of whether the reaction was initiated by ultraviolet light or by ^{60}Co gamma radiation.

These similar product mixtures can be accounted for by assuming that the chain-propagating and chain-terminating steps of the reaction are the same, although the initiation steps may differ in detail. With chlorine and with toluene or benzene in the reaction mixture, ultraviolet light is only energetic enough to dissociate the chlorine molecules present. However, absorption of ionizing radiation by the reactants can be expected to cause excitation and ionization of all species present. Similarities in the product mixtures from the ultraviolet-radiation-initiated reactions indicate that the ionic species created by ionizing radiation probably do not react as such but are first neutralized, yielding excited species which then react in chain-propagating steps similar to those of the excited species formed in the light-initiated reaction. Thus the first discernible chemical reaction in the ionizing-radiation-initiated reaction may be addition of a chlorine to an aromatic ring or removal of a hydrogen from a side chain.

In the most probable mechanism for the free-radical addition-substitution reaction, the initiation step with either photochemical or ionizing-radiation activation is

$$Cl_2 \rightsquigarrow 2Cl\cdot$$

In addition to this step, ionizing radiation can, in the presence of benzene, cause

$$C_6H_6 \rightsquigarrow C_6H_6^*$$

$$C_6H_6^* + Cl_2 \rightarrow C_6H_6 + 2Cl\cdot$$

and/or

$$C_6H_6^* + Cl_2 \rightarrow C_6H_6Cl + Cl\cdot$$

and, with toluene or other alkylaromatics,

$$C_6H_5CH_3 \rightsquigarrow C_6H_5CH_3^*$$

$$C_6H_5CH_3^* + Cl_2 \rightarrow C_6H_5CH_3 + 2Cl\cdot$$

and/or

$$C_6H_5CH_3^* + Cl_2 \rightarrow C_6H_5CH_2\cdot + HCl + Cl\cdot$$

and/or

$$C_6H_5CH_3^* + Cl_2 \rightarrow C_6H_5ClCH_3 + Cl\cdot$$

Chain-propagating steps common to both the ultraviolet and ionizing-radiation-initiated reactions in the addition of chlorine to the aromatic ring can be represented as

$$C_6H_6 + Cl\cdot \rightarrow C_6H_6Cl\cdot$$

$$C_6H_6Cl\cdot + Cl_2 \rightarrow C_6H_6Cl_2 + Cl\cdot$$

$$C_6H_6Cl_2 + Cl\cdot \rightarrow C_6H_6Cl_3\cdot$$

$$C_6H_6Cl_3\cdot + Cl_2 \rightarrow C_6H_6Cl_4 + Cl\cdot$$

$$C_6H_6Cl_4\cdot + Cl\cdot \rightarrow C_6H_6Cl_5\cdot$$

$$C_6H_6Cl_5\cdot + Cl_2 \rightarrow C_6H_6Cl_6 + Cl\cdot$$

and those for alkyl substitution as

$$C_6H_5CH_3 + Cl\cdot \rightarrow C_6H_5CH_2\cdot + HCl$$

$$C_6H_5CH_2\cdot + Cl_2 \rightarrow C_6H_5CH_2Cl + Cl\cdot$$

etc.

The common chain-terminating steps can be represented as

$$Cl\cdot + Cl\cdot \rightarrow Cl_2$$

$$C_6H_6Cl\cdot + Cl\cdot \rightarrow C_6H_6Cl_2$$

$$C_6H_6Cl_3\cdot + Cl\cdot \rightarrow C_6H_6Cl_4$$

$$C_6H_6Cl_5\cdot + Cl\cdot \rightarrow C_6H_6Cl_6$$

$$C_6H_5CH_2\cdot + Cl\cdot \rightarrow C_6H_5CH_2Cl$$

etc.

In the free-radical chlorination of aliphatic compounds, the ease of hydrogen displacement by the attacking chlorine atom is well established to be tertiary > secondary > primary.[3] This selectivity is increased in the vapor phase and decreased as the temperature is increased in the liquid phase. These effects are explained as being the inverse of the order of the C–H dissociation energies (primary > secondary > tertiary) and reflect the resonance stability of the resulting intermediate radical. However, the alkyl groups of alkylaromatic compounds do not conform to this simple energetic picture. Thus, in photochemical chlorination, C–H bonds of the methyl group of toluene are less reactive toward abstraction of a hydrogen by chlorine (by a factor of 2.7) than those of cyclohexane, despite the great resonance stability of the benzyl radical.[56] The presence of a chlorine substituent on the side chain further decreases its reactivity, so that xylene will preferentially form α,α'-di(chloromethyl) benzene. The selectivity of chlorine atom attack on the side chain, e.g., at the primary vs. tertiary position in cumene, is reported to depend on the total concentration of the aromatic molecules present.[57] From these data it has been concluded[57,58] that the transition state in the free-radical substitutive chlorination of alkyl substituents on an aromatic ring involves little breaking of the C–H bond (to explain the lack of effect of potential resonance stabilization) and that the reaction may preferentially involve chlorine atoms or molecules that have formed a π complex[59] with the aromatic ring (to explain the concentration effects). The π complex is probably also involved in the addition reaction. If the chlorine atom adds to a previously formed benzene–chlorine complex, the addition of the second chlorine — one of the chain-propagating steps — would be greatly facilitated.

LET Effect on Chlorination

Different sources of ionizing radiation may interact within liquids in different manners as a result of different (local) ionization densities. Gamma radiation causes ionization and excitations to occur relatively far apart from one another, so that the individual ionized or excited molecule has a high probability of interacting with other molecules rather than with other ionized or excited species in the medium. On the other hand, more densely ionizing alpha radiation causes ionizations and excitations to occur sufficiently close together that the reactive species can and do react with each other as well as within the bulk medium. This effect of the different types of incident radiation on the medium on which they are acting is measured by the LET. The LET is defined as the differential rate of loss of energy per unit length of track of the ionizing particle. The values of the LET increase with the square of the charge on the particle and decrease as the speed of the particle increases.[60–62] Typical values of the LET for water are given in Table 1.

Very little is known about the effects of LET in the chlorination of aromatics. The reported G values for the chlorination of benzene using several different types of radiation are given in Table 2. Before firm conclusions can be drawn from this table, however, it must be pointed out that a substantial dose-rate dependence is observed for this reaction[22] so that, lacking data at comparable dose rates, the LET effect may be obscured by the overall dose-rate dependence. Even with similar dose rates, different G values have been reported by different researchers.[22,23] It can be seen, however, that the reaction initiated by beta radiation from a ^{32}P source[25] appears to have a smaller yield (G value) than does that initiated by a ^{60}Co source of similar dose rates;[23] i.e., the higher LET radiation results in a lower yield. No information is available on the comparability of the reaction conditions (e.g., concentration, temperature) in these two experiments. However, in experiments that closely duplicated all reaction conditions,[22] a slightly higher G value was observed when the gamma-radiation source was changed from ^{60}Co to ^{137}Cs (which emits gamma radiation of lower energy). The G value for the reaction initiated by

Table 1 MEAN LET VALUES FOR WATER[60–62]

Radiation	LET, kev/μ
From uranium fission fragments	4000
1-Mev alpha	560
10-Mev alpha	120
Polonium-210 alpha (5.3 Mev)	150
1-Mev proton	60
10-Mev proton	10
0.01-Mev electron	4.9
0.1-Mev electron	0.8
1.0-Mev electron	0.4
Cobalt-60 gamma (1.25 Mev)	0.42
200-kev X	2.8
20-Mev X	0.28

Table 2 G VALUES FOR THE CHLORINATION OF BENZENE

Dose rate, ev/(g)(min)	G value	Radiation	Chemical reactor type	Ref.
Not reported	2,160	α (^{222}Rn)	Batch	19, 63
1.79×10^{15}	380,000	β (^{32}P)	Batch	25
2.34×10^{15}	790,000	γ (^{60m}Co)	Batch	23
4.10×10^{15}	560,000	γ (^{60}Co)	Batch	23
5.60×10^{15}	370,000	γ (^{60}Co)	Batch	23
8.90×10^{15}	210,000	γ (^{60}Co)	Batch	25
2.11×10^{16}	180,000	γ (^{60}Co)	Batch	23
4.39×10^{16}	43,000	γ (^{60}Co)	Continuous	22
6.17×10^{16}	84,000*	γ (^{60}Co)	Batch	20, 21
6.40×10^{16}	61,000	γ (^{60}Co)	Continuous	22
2.10×10^{17}	20,000	γ (^{60}Co)	Continuous	22
3.00×10^{17}	22,000	γ (^{60}Co)	Continuous	22
3.15×10^{17}	34,000	γ (^{137}Cs)	Continuous	22

*For a reaction mixture of 20% benzene in carbon tetrachloride.

alpha radiation (from ^{222}Rn) is the lowest reported,[19,60] further indicating an inverse relation between LET and yield (G value) in the chlorination of benzene.

In the radiolysis of the pure aromatics, substantial increases in G values with increasing LET have been reported. The G values for loss of starting materials as well as those for the many gaseous, liquid, and solid products increase when radiation of higher LET is used with both benzene[64] and toluene[65] as target materials. Thus the use of more densely ionizing forms of radiation (i.e., radiation with higher LET) to initiate the chlorination reaction on aromatics would lead to more undesirable by-products resulting from direct radiolysis of the aromatic substrates as well as probably decreasing the yields of the desired products.

Potential Radiation Sources

The radiation for initiation of the chlorination reaction can be obtained from either an external or an internal radiation source. External sources have been used for all the ionizing-radiation-initiated chlorinations discussed above. Most of these processes have utilized gamma-emitting radioisotopes such as ^{60}Co (Refs. 15, 20 to 25, 45, 47, and 49) and ^{137}Cs (Ref. 22), although some use has been made of machine (electron) irradiators.[47] Either of these types of sources requires extensive shielding and remote operation of the reaction vessel. Several irradiator designs for remote operation of the chlorination process have been proposed,[21,22] incorporating either flat plaque sources or a single source in a center well.

Placing the radiation source within the reaction vessel would better distribute the source of radiation among the reacting molecules and permit a more efficient utilization of the emitted radiation. Either gamma- or beta-emitting radioisotopes could be used for such an internal source. The properties of a number of possible candidate isotopes have recently been tabulated.[66] However, an internal beta source would afford a high level of radiation in the reaction zone for a relatively smaller source.[67] The beta rays deposit the bulk of their energy along a short path within the reaction vessel, where the energy can most effectively be used. Gamma rays dissipate energy over a longer range with a smaller fraction of the energy being deposited within a given reaction vessel. A larger reaction vessel would be required to better use the energy given off by a gamma-emitting source. However, other considerations, such as efficient and sufficient mixing of the reactants, may preclude a reactor design that would make the best use of gamma radiation. For these reasons a beta-emitting source may be preferred. Of the possible isotopes,[66] only ^{90}Sr in equilibrium with its daughter, ^{90}Y, provides a beta source with suitable half-life for commercial utilization with minimum shielding requirements.

The internal beta source may be used in a variety of shapes. The least efficient, in terms of energy available for utilization, is a flat plaque source affixed to a wall or bottom.[67] Better energy utilization is possible by any one of several methods currently or potentially available which present large surface areas of the contained source to the reactants.

Small spherical sources (trade named Microspheres) are commercially available from the Minnesota Mining and Manufacturing Company.[68] These sources are in the form of ceramic spheres with a softening point above 1500°C. They are claimed to have high chemical, physical, and radiation stability. One hundred curies of ^{90}Sr as Microspheres gives a dose rate of 1 Mr/hr. These sources are available with or without secondary encapsulation and in a variety of sphere sizes. A prototype source incorporating Microspheres loosely packed inside thin-walled stainless-steel needles has been used for a study of the effect of radiation on the hydrogenation of coal and coal distillates.[69] This 90-curie source had an irradiation efficiency of 34.7% and produced an effective dose rate of 9.8×10^5 r/hr. Unfortunately, under the high temperature and pressure conditions of this work (430°C at 5000 psi), considerable leaching of the Microspheres was observed following the formation of cracks in the stainless-steel cladding. Further secondary encapsulation (for example, by use of a thicker wall) would lessen this problem but at the expense of irradiation efficiency.

Work at the Oak Ridge National Laboratory on the production of several glasslike sources has been reviewed by Lewis.[70] Various silicates, including those with boron and aluminum, constitute these glasses. Aluminosilicate glasses are produced by absorbing the radioactive material on an aluminosilicate ion exchanger (Decalso), mixing with a flux such as calcium oxide, and fusing at high temperature. These sources are reported to have good abrasion resistance, minimizing the attrition problem. The radioactive materials thus treated include ^{137}Cs, ^{90}Sr, ^{144}Ce, ^{241}Am, ^{204}Tl, and ^{233}U.

Silicate and borosilicate glasses have been investigated,[70] primarily with respect to the fixation of ^{137}Cs. In these studies the cesium becomes an integral part of the glass, resulting in production of either a cesium silicate or a cesium borosilicate glass. These glasses have good resistance to leaching and volatilization. The cesium borosilicate glass has been investigated as a possible isotopic power source.[71] A similar glass incorporating ^{90}Sr to form a strontium silicate or strontium borosilicate would result in a potentially suitable source. The form of this source could be varied. Thus a pebble or spherical source could be relatively easy to produce from the glass.

Compressed strontium titanate pellets have good thermal and radiation stability with low leaching rates even without encapsulation.[66] Thin-walled encapsulation of small sintered pebbles or beads would

thus produce a potential source. To date, however, no isotopic source of this composition or configuration has been tested.

Another form considered potentially useful as an internal beta source is that of helix-wound tubing. Sources of this type are commercially available from Beta Radiation, Inc.[72] They are fabricated by swaging a metallic tube containing the source material, generally sintered strontium titanate, down to a small diameter using standard swaging equipment, thus compressing and compacting the source material. The tubing is generally of stainless steel with a 5-mil wall thickness. The wall thickness is not changed by the swaging process.

A source of this type was used very successfully in studies of hydrogenation of coal distillates[73] and in some vapor-phase telomerization studies.[74] This source, fabricated to contain 400 curies of ^{90}Sr (as strontium titanate) was 9 ft long with a diameter of 0.077 in. For use in these experiments, the tubing was wound into a helix of 1 in. diameter and 4 in. height. In this configuration the radiation efficiency was 22.8%. An effective dose rate of 1.69×10^6 r/hr was reported.[73] This 400-curie source required only 1 in. of lead for shielding. Even under the high temperature and pressure used (up to 500°C at 5000 psi), no radioactive contamination of the reactant was observed.[73,74]

It thus appears that several possibilities exist for using a ^{90}Sr beta source within the reaction vessel to initiate the chlorination reaction. The major problem to be overcome is the possible attrition with an unencapsulated source such as Microspheres or one of the glasses. Use of an unencapsulated source would, of course, be preferred since there is less energy loss due to self-absorption. However, thin-walled stainless-steel encapsulation, such as on the wire sources from Beta Radiation, Inc., prevents attrition with only a small loss in radiation efficiency. (MG)

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Section

IV

Isotopes and Radiation Technology

Life Sciences

The Anger Camera

By Joan E. Carden

Abstract: Isotope or organ visualization with a radioisotope scintillation camera—an instrument which has a detector that is fixed in relation to the patient and which produces pictures called images—is a diagnostic technique for determining the localization of a small quantity of gamma-ray or positron-emitting isotope administered to a patient. A specific radioisotope camera, the Anger camera developed by Hal O. Anger, University of California, Berkeley, is described here in detail. The description of the camera is followed by citations of some selected instances in which the Anger camera has been used.

This paper is a selective review of the literature on the Anger radioisotope camera.* Radioisotope scanning is a diagnostic technique for visualizing the localization of a small quantity of gamma-ray- or positron-emitting isotope administered to a patient.[1-3] The localization of a radioactive tracer in the body depends on the chemical nature of the radioactive material and the biological and biochemical functions of the organs or areas under consideration. Thus a different concentration of radioactive material may be accumulated in an organ than in the surrounding tissues, or regions *within* an organ may contain different concentrations of radioactive material due to impaired function.

The two principal means of visualizing the localization of a radioisotope are (1) the scanner, which has a detector that moves in relation to the patient and produces pictures called "scans," and (2) the scintillation camera, which has a detector that is fixed in relation to the patient and produces pictures called "images."

Modern radioisotope cameras have several advantages over radioisotope mechanical scanners:[4]

1. They take pictures much more rapidly. The speed at which they take successive pictures is limited only by the rate at which radiation is detected, and not by any mechanical factors. Brain-tumor pictures have been taken in 5 sec, and stop-motion pictures of ^{99m}Tc going through the heart have been taken at a rate of one picture per second. Scanners require at least several minutes to scan most organs since their speed is limited by how fast their probes can travel over the area being scanned.

2. The best currently available radioisotope cameras are 5 to 50 times as sensitive as conventional scanners. This sensitivity is achieved partly because they view the entire picture area continuously (at present the area is limited to 10 in. diameter), while scanners view only one point at a time.

3. The shorter exposure times encourage taking views of organs from different angles; i.e., the patient can be sitting, standing, or lying down, and the camera positioned to obtain frontal, lateral, or oblique views under any conditions, thereby improving the visualization of lesions and other abnormalities with respect to natural anatomical relationships.

4. As a direct result of shorter exposure times, rapid sequences of still pictures and motion pictures can be taken to illustrate the movement of tracer compounds through organs. Numerical count rate data can be obtained as a function of time for specific picture areas. For example, curves of count rate vs. time can be obtained for a particular part of the kidney or a chamber of the heart. The ability to observe changes in the distribution of compounds in animal and human subjects and to quantitate the uptake and movement of the compounds as a function of time provides a powerful new method of medical investigation.

5. More patients per day can be studied, which is an important economic advantage. The camera is readily adapted to the patient in a natural and comfortable position, but the patient usually must be "adjusted" to mechanical scanners.

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The principal disadvantages of the scintillation camera compared to mechanical scanners stem from limitations:

1. On the thickness of the scintillation crystal. On the one hand, if the crystal thickness is > 0.5 in., a significant loss of resolution occurs because of the increased probability of a single gamma ray undergoing multiple interaction. Although camera-type instruments, including the Anger camera, do not give better resolution than mechanical scanners,[5] they do give better depth of focus; i.e., they maintain their resolution over a greater range of depth than scanners employing large crystals. The thin-crystal requirement results in a decrease in the efficiency that is especially important when detecting >300-kev gamma rays.

2. On the diameter of the scintillation crystal which limits the size of field obtainable. Present instruments view a 10-in.-diameter area, and future instruments will probably have a 14-in. field.

Whereas radioisotope cameras were briefly discussed in a previous issue of *Isotopes and Radiation Technology*,[6] the present discussion is a detailed description of a specific camera, the Anger radioisotope camera, developed by Hal O. Anger.* The description of the Anger camera is followed by selected instances in which the camera has been used.

be refracted or focused and photographed in the usual sense, an alternative is the method of selective interference—blocking out or absorbing all but a small fraction of the rays with collimators. The collimator may be of the pinhole type, consisting usually of a hole in a conical lead shield, or of the multichannel type, consisting of hundreds or thousands of channels in an absorber plate. An image of the subject can be obtained if a radiographic film or other detection means is located on the opposite side of the collimator.

In 1896, Roentgen used a pinhole aperture and photographic film to obtain an image of the X-ray-emitting anode of one of his first tubes. Copeland and Benjamin,[7] in 1949, used a gamma-ray pinhole camera and X-ray film to obtain a picture of a 6-mc radium needle—with an exposure time of 24 hr! In 1952, Anger[8] used a gamma-ray pinhole camera, a solid NaI(Tl) crystal to convert gamma rays to visible light, and a blue-sensitive photographic plate in close proximity to the NaI scintillation crystal to obtain pictures of a metastatic thyroid lesion in vivo. An ^{131}I activity of 1 mc/cm^2 and a 1-hr exposure were necessary to obtain detectable blackening of the film.

The sensitivities of the above techniques were obviously too low for clinical diagnostic use. It would be of obvious interest in tracer turnover studies and in medical diagnostic work to have devices that would

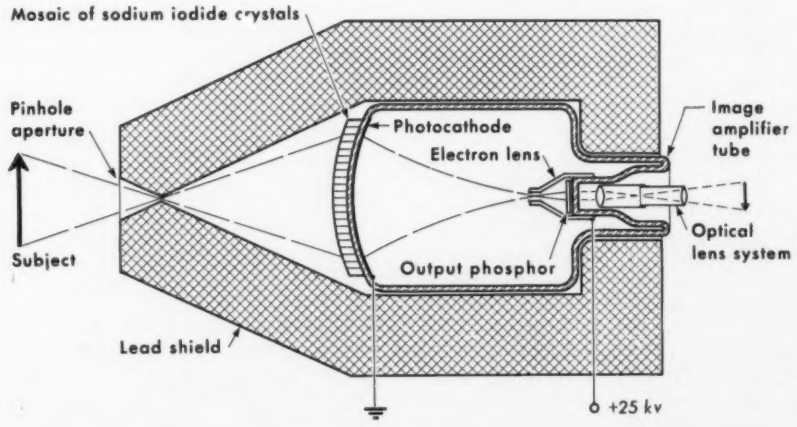


Fig. 1 Early version of gamma-ray camera with an image-amplifier tube.

Early Radioisotope Cameras

Of the various particles and rays emitted by radioactive isotopes, only X rays, gamma rays, and photons generated incidental to positron decay have enough penetrating power to reach the surface of the body from deep-lying organs. Although these rays cannot

show the accurate in vivo distribution of radioactive substances in the form of visual images, at a level where the administered dose of radioactivity would have negligible biological effect and at an exposure time suitable for studies involving dynamic systems. In 1954, Mortimer, Anger, and Tobias[4,9] developed a method of converting the gamma-ray image to a visible-light image by a scintillator and then amplifying the light with an image intensifier (Fig. 1). To obtain an image of activity distribution, a collimator

*Donner Laboratory, University of California, Berkeley.

first projects a gamma-ray image of the subject on a mosaic of 241 NaI(Tl) scintillation crystals. The ionizing radiation is absorbed by the scintillator, and light is emitted. A very sensitive detector of light is necessary[10] to obtain a record of the absorption by the crystal of an individual gamma ray. (Such a single-stage image-amplifier tube was obtained from Philips Research Laboratories.*) The light emitted from the scintillator falls on the 5-in.-diameter photocathode of the image amplifier, and electrons are produced. When a 25-kv positive voltage is applied, these electrons are accelerated (although not increased in number as in a multiplier phototube) within the image amplifier. The electrons are focused by an electron lens onto a fluorescent screen, and the intensified [by a factor of 600 (Ref. 8)] image that appears on the output screen is viewed directly or recorded on photographic film. Despite the fact that the sensitivity of this instrument leaves much to be desired, this method of using image intensifiers in radioisotope cameras demonstrated the potential usefulness of camera-type imaging devices.

In 1956, 2 years after the above radioisotope camera had demonstrated the potential usefulness of camera-type imaging devices, Anger developed his "scintillation" camera.[4,11,12] He abandoned the image-amplifier tube as a detector of light and used multiplier phototubes instead. In a multiplier phototube a photocathode receives light emitted from the scintillation crystal, the electrons emitted by the photocathode are attracted to the anode of a dynode, and for each electron received several electrons are emitted from the dynode and are, in turn, attracted to the anode of another dynode. Within one multiplier phototube there may be 10, 12, or even 16 dynodes, and $\sim 10^6$-fold amplifications of pulses of current thereby result. The size of the pulse that emerges from these phototubes is approximately proportional to the energy of the absorbed radiation. This relation between energy and pulse height is subject to statistical fluctuation.

In the scintillation camera (Fig. 2), some of the gamma rays emitted from a subject travel through the aperture in the lead shield and continue traveling in straight lines until they impinge on the 4-in.-diameter by 0.25-in.-thick, solid NaI(Tl) scintillation crystal (solid as opposed to mosaic because the solid images have no distracting patterns superimposed). The locations of scintillations occurring in the crystal are determined from the amount of light picked up by seven 1.5-in.-diameter multiplier phototubes simultaneously viewing the crystal. The light produced in any given scintillation is emitted isotropically and thus is divided among all the phototubes, with those closest to a given scintillation receiving the most light.

*Philips Gloeilampenfabrieken, N. V. Forschungslaboratorium, Eindhoven, Netherlands.

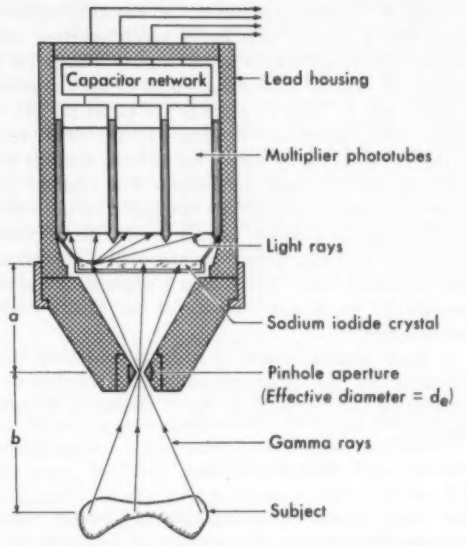


Fig. 2 The "scintillation" camera of 1956 with seven multiplier phototubes.

The circuit for computing the location of scintillations and displaying them on the image-readout oscilloscope is shown in Fig. 3. The pulses from the phototubes are fed through small capacitors to four output leads called the X^+, X^-, Y^+, and Y^- leads. The amount of signal transferred to each output lead is proportional to the capacitance in picofarads (see Fig. 3). For instance, phototube No. 1 has a 30-pf capacitance to the X^+ lead, 10 pf to the X^- lead, 40 pf to the Y^+ lead, and zero capacitance to the Y^- lead. Therefore this phototube transfers most of its signal to the X^+ and Y^+ leads. If a scintillation occurs directly under tube No. 2, most of the isotropically emitted light is collected by tube No. 2 because this tube is closest and subtends a large solid angle. Smaller amounts are collected by tubes Nos. 1, 3, and 7 and still smaller amounts by tubes Nos. 4, 5, and 6. A large X^+ signal is generated by this scintillation because tube No. 2 is connected by a large capacitance (40 pf) to the X^+ lead. A small X^- signal is generated, and moderate but equal Y^+ and Y^- signals are generated.

The X^+, X^-, Y^+, and Y^- signals are applied to analog computer elements, and three new signals are produced. Two are positioning signals approximately proportional to the X and Y coordinates of the scintillation and are obtained from:

$$X = X^+ - X^-$$
$$Y = Y^+ - Y^-$$

Continuing with the scintillation occurring directly under phototube No. 2, we find that, when the X^+, X^-,

Y^+, and Y^- signals are applied to the difference circuits, the X circuit puts out a large positive signal because the amplitude of the X^- signal is large (40 pf), and the Y circuit puts out a signal of zero amplitude because the Y^+ and Y^- signals are equal (20 pf). The origin of the coordinate system is located at the center of the crystal. These equations assume that all scintillations are of equal brightness. For reasons given below, the only scintillations displayed on the readout oscilloscope are those nearly equal in brightness (photopeak scintillations) so the above condition is approximately met. The X and Y signals are applied directly to the X and Y inputs of the image-readout oscilloscope.

A third signal, which is proportional only to the *brightness* of the scintillation without regard to its location in the crystal, is the Z signal. It is calculated by:

$$Z = X^+ + X^- + Y^+ + Y^-$$

This signal is applied to the input of a single-channel pulse-height selector. The window of the pulse-height selector is adjusted to accept only the photopeak of the gamma-ray spectrum; thus most scattered gamma rays and many due to natural background are eliminated because they do not meet the pulse-height requirement.

When the X and Y signals are applied (i.e., when a scintillation occurs), the image-readout oscilloscope beam, which is normally directed to the center of the screen, is deflected to a point on the right-hand side of the screen for 1 μsec. At the same time the X^+, X^-, Y^+, and Y^- signals have been summed in the addition circuit and the resulting Z signal applied to the input of the pulse-height selector. If the Z signal has the prescribed amplitude, the pulse-height selector produces an output pulse that turns on the beam of the image-readout oscilloscope for 0.5 μsec. The scintillation is reproduced as a point flash of light on the right-hand side of the oscilloscope screen. The flashes are photographed over a period of time with an oscilloscope camera, and the resulting picture shows the distribution of activity in the subject. Exposure times last from 1 sec to a few minutes or more, and usually 1 thousand to 100 thousand or more dots are recorded on each picture. Although only seven phototubes are used in this model of the scintillation camera, hundreds of picture elements can be resolved because the scintillations are reproduced in approximately correct locations even when they occur at points intermediate between the phototubes. For imaging large subjects, such as the brain or liver, for the distribution of a gamma-ray isotope, multichannel collimators give the best combination of sensitivity and resolution.[13] However, for small gamma-ray-emitting subjects, such as the thyroid gland, pinhole collimation is the method of choice. Coincidence collimation is used to image both large and small subjects with positron-emitting isotopes.

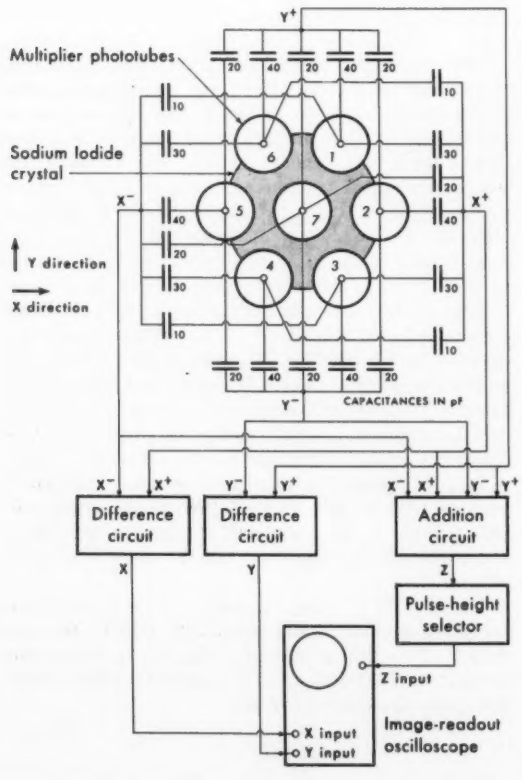


Fig. 3. Block diagram showing principle of operation of 1956 "scintillation" camera with seven multiplier phototubes.

The 1967 Anger Radioisotope Camera

The availability of larger NaI crystals has enabled the construction of a camera with higher sensitivity and better overall resolution than that of the earlier models. The current version of the Anger camera* (Fig. 4)—currently the most widely used of clinical radioisotope cameras—is a modified version of the 1956 scintillation camera.[12,14]

*Pho/Gamma III manufactured by Nuclear-Chicago Corporation. Several accessories, such as the positron attachment, 35-mm time-lapse camera, and chart recorder, are available. A complete Model 6403 Pho/Gamma III system, including a scintiphoto copying system for use with Polaroid film and three collimators—a single pinhole collimator, a 3-in.-thick 1000-hole collimator, and a 1.75-in.-thick 4000-hole collimator—is catalog priced at $34,440.00 (f.o.b. factory). A complete system for use with positron emitters, Model 6410 Positron III, is priced at $47,125.00 (f.o.b. factory).

Fig. 4 Commercially manufactured Anger scintillation camera (Nuclear-Chicago's Pho/Gamma III Scintillation Camera). Gamma-detector specifications include an 11.5-in.-diameter by 0.5-in.-thick NaI(Tl) crystal and nineteen 3-in.-diameter multiplier phototubes. (Photograph courtesy of Nuclear-Chicago Corporation.)

In the Anger camera image detector (Fig. 5), the 11.5-in.-diameter by 0.5-in.-thick solid NaI(Tl) scintillation crystal, which is roughened on all sides to decrease piping of the light to the edges of the crystal by repeated internal reflection, is contained in a hermetically sealed can with a glass window and an aluminum oxide reflector. The scintillation crystal is reviewed by a hexagonal array of nineteen 3-in.-diameter multiplier phototubes. An important component that has been found empirically to improve the resolution and linearity of the pictures is a light deflector adjacent to the phototubes to reflect light that would normally fall between the tubes.

The electronic circuit, including the extra components for coincidence collimation* of annihilation radiation from positron emitters, is diagrammed in Fig. 6. Basically, the principles of operating this camera and the 1956 scintillation camera are the same. The gamma-ray energy selector permits one to adjust the pulse-height window to an isotope of given gamma-ray energy while the phototube voltages are kept constant. The Z-pulse monitor oscilloscope provides a means of quickly adjusting the pulse-height selector window to the photopeak of the isotope in use. The capacitor network is justifiably more complicated because of the larger number of phototubes used. And the difference circuits have been replaced by ratio circuits,

$$X = (X^+ - X^-)/Z \text{ and } Y = (Y^+ - Y^-)/Z$$

so that the positions of flashes on the image-readout oscilloscope are independent of the brightness of the scintillation. Wide pulse-height selector windows can therefore be used without the slight loss of position resolution at the edges of the pictures of the difference-circuit instruments.

The useful gamma-ray energy range of a phototube array scintillation camera is approximately 70 to 700 kev.†

Applications

The Anger radioisotope cameras have been used in numerous studies,[15] including those on the liver with ^{131}I-labeled Rose Bengal; brain tumors with ^{203}Hg, the positron emitter ^{68}Ga, and ^{99m}Tc; the heart with ^{99m}Tc; the thyroid with ^{131}I; bone tumors and bone-marrow distribution with the positron emitters ^{18}F and ^{52}Fe, respectively; dynamic blood flow with ^{99m}Tc; and intestinal iron absorption with ^{52}Fe.

In the visualization of blood flow in the brain and kidneys, by Powell and Anger,[16] a significant achievement — dynamic studies — of the radioisotope camera is demonstrated. In this study, 10 mc of ^{99m}Tc, which is obtained from a ^{99}Mo generator, is administered as the pertechnetate into an antecubital vein (i.e., in the inner or front surface of the forearm). The pulse-height selector of the camera is set for 0.125 to 0.155 Mev, and a thin-septum multichannel collimator is used. The camera is switched on 5 sec after the injection, and the Polaroid roll film is pulled out manually, one frame every 5 sec with the Polaroid camera shutter open continously. A series of eight 5-sec ex-

*Whenever a positron is stopped by matter, it combines with a negative electron and produces two 0.51-Mev annihilation gamma rays that travel from their point of origin at 180° with respect to each other. This property, along with coincidence circuitry and electronic computation techniques, allows images to be formed with two detectors on opposite sides of the subject. The remote gamma-ray counter is called a focal detector because all gamma-ray pairs that form an image of the subject must impinge on it, and it therefore is a focal point in the image geometry. Conventional image-producing collimators, such as pinhole or multichannel collimators, are not required. The positron camera in Fig. 6 is one of several types of such cameras, which vary from each other in detection efficiency, sensitivity, and resolution.[12] The types include those with:

1. Two identical image detectors [two 0.5-in.-thick solid NaI(Tl) crystal image detectors]
2. Two dissimilar image detectors [a 0.5-in.-thick solid NaI(Tl) crystal image detector and a large scintillation counter with a thick NaI(Tl) crystal as a focal detector]
3. Image detector [0.5-in.-thick solid NaI(Tl) crystal] and focal detector [a single, large, thick NaI(Tl) crystal viewed by an array of multiplier phototubes (Fig. 6)]
4. Image detector [< 0.5-in.-thick solid NaI(Tl) crystal] and focal detector [array of scintillation counters individually equipped with a thick NaI(Tl) crystal]

†Therefore other radioisotope cameras have evolved, such as the two-stage image-amplifier scintillation camera system developed by Ter-Pogossian[4, 12, 17] for visualization of organs containing low-energy X- and gamma-ray-emitting isotopes (< 150 kev). Beierwaltes[18] is currently developing a multistage image-intensifier scintillation camera that can rapidly record the radioisotope distribution in the body with very high resolution (1 mm). Other radioisotope cameras include the autofluoroscope developed by Bender and Blau[19] and the spark-chamber gamma-ray camera by Lansiart and Kellershohn.[15]

posures on Polaroid film and 1-sec exposures with a 16-mm movie camera is made simultaneously.

The cerebral blood flow is imaged with the camera viewing the anterior aspect of the head, and the renal blood flow, with the camera viewing the posterior aspect of the kidneys. Figure 7 shows 5-sec-exposure Polaroid pictures for a patient with *(a)* normal abdominal aortic blood flow and renal perfusion and *(b)* a large left renal cyst, the outline of which is readily identified in the lateral margin of the left kidney as a dark area that is void of perfusion by ^{99m}Tc. Whereas the conventional indirect methods of estimating renal blood flow are accompanied by difficulties of interpretation, the Anger camera enables a rapid and safe estimate of blood flow and tissue perfusion. The greater complexity of cerebral circulation makes interpretation of cerebral perfusion pictures more complicated, but the Anger camera provides estimation of tissue perfusion not visualized by arteriograms.

In intestinal studies it is known[20] that, following an oral dose of iron, absorption from the intestinal tract occurs in two phases: an early, rapid phase that occurs within 2 hr and accounts for 40 to 60% of the total dose absorbed; and a late, slow phase that occurs after 24 to 48 hr. Whereas the upper small intestinal tract, particularly the duodenum, had been credited as being responsible for the rapid phase, the site accounting for the slow phase had not been clearly defined.

Fawwaz et al.[20] orally administered to fasting normal subjects 40 μc of the positron emitter ^{52}Fe as ferrous chloride with 4 mg of carrier ferrous sulfate. Before the oral dose and after the conclusion of the experiment, iron-turnover studies are performed with 2 μc of transferrin-bound ^{59}Fe injected intravenously.

Using the Anger positron camera, photoscans (Fig. 8) of the abdomen of a normal subject are taken throughout the study, and the ^{52}Fe content of the plasma of blood samples taken at 0.5-hr intervals is determined with a NaI(Tl) scintillation counter. The bar graph at the top of each photoscan in Fig. 8 represents the calculated percentage of ^{52}Fe absorbed into the plasma at 0.5-hr intervals. (The rate of ^{52}Fe absorption into the plasma at any given time is the sum

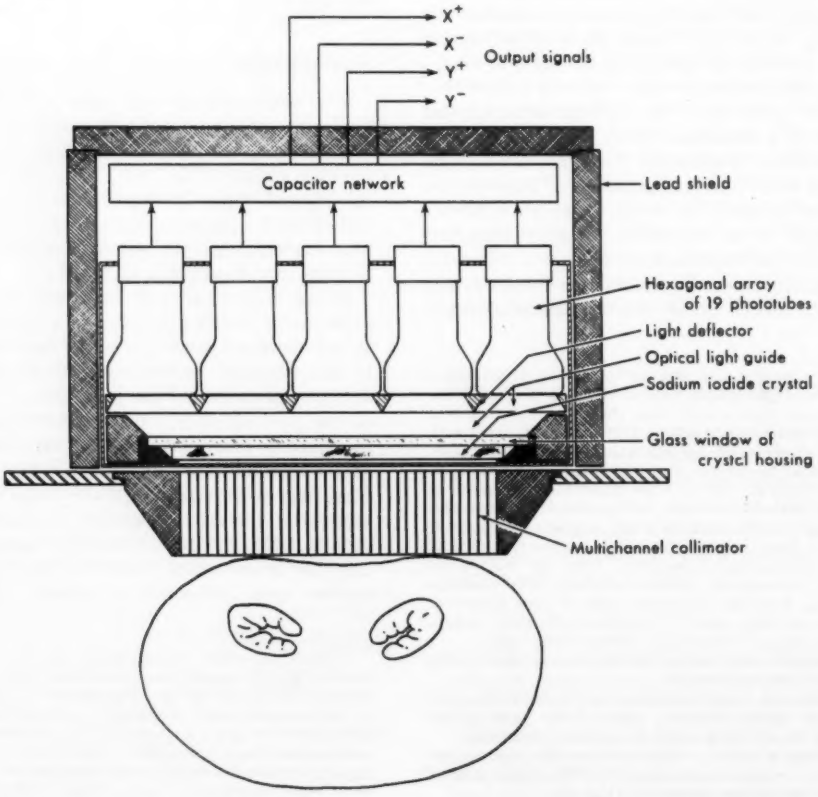


Fig. 5 Image detector of Anger scintillation camera with 11.5-in.-diameter, 0.5-in.-thick solid NaI(Tl) crystal and 19 multiplier phototubes. Multichannel collimator is shown between crystal and subject.

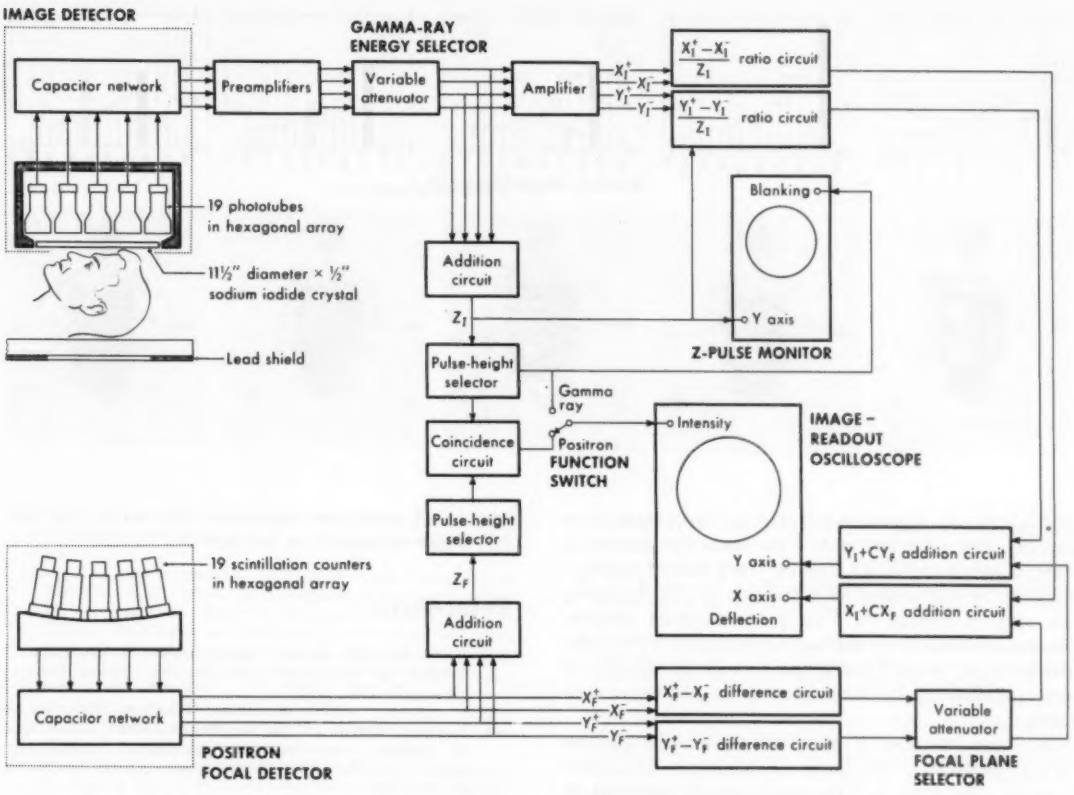


Fig. 6 Block diagram of gamma-ray and positron scintillation camera with image detector shown in Fig. 5.

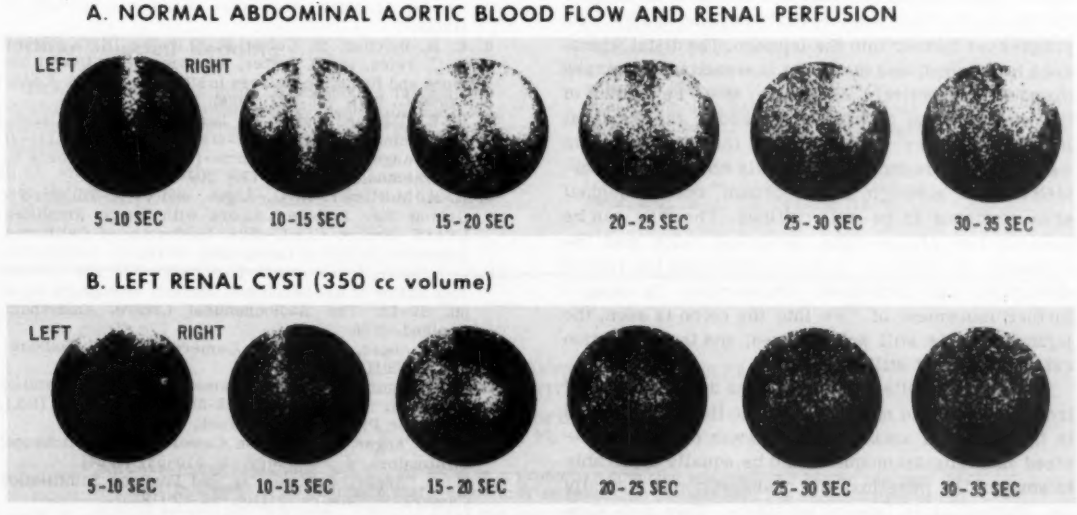
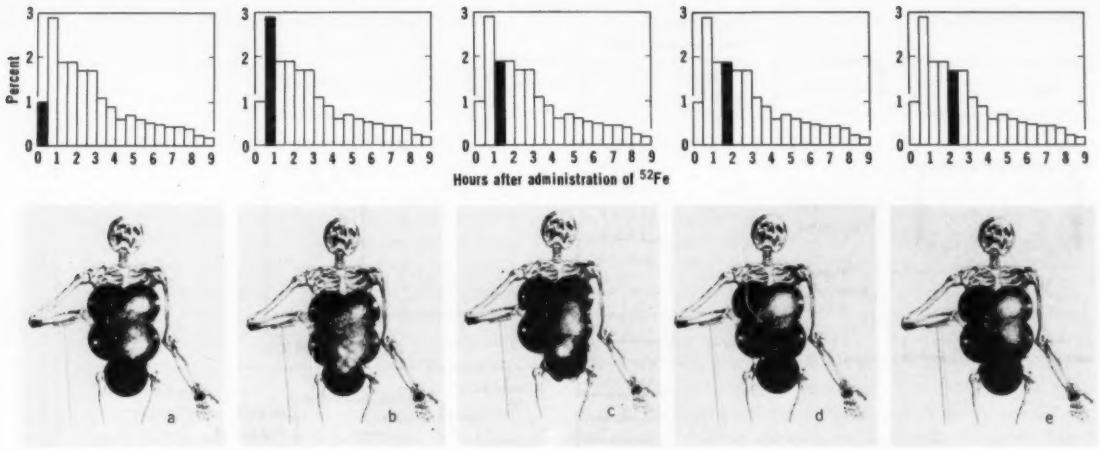


Fig. 7 Six 5-sec Polaroid posterior-view exposures of (a) *normal and* (b) *abnormal kidneys after injection of ^{99m}Tc in the bloodstream.*



of the rate of change in the plasma ^{52}Fe at that time and the rate of removal of ^{52}Fe from the plasma to bone marrow and other tissues. This rate of absorption can be calculated as a function of time by using the rate of change of ^{52}Fe plasma levels and the plasma iron-turnover rate as measured by ^{59}Fe.) The shaded area of each bar graph represents the rate of absorption during the time interval the photoscan was taken.

In image *a* of Fig. 8, absorption has begun and the ^{52}Fe is present in the stomach duodenum and upper jejunum. The rate of ^{52}Fe absorption is maximal in image *b*, where progress of the ^{52}Fe into the jejunum is seen; this image demonstrates that, at the time of maximal ^{52}Fe absorption, the ^{52}Fe is present in the upper small intestinal tract and stomach. In image *c* the absorption rate has started to decline, and the ^{52}Fe progresses further into the jejunum. The distal jejunal area is reached, and the ileum is visualized in images *d* and *e*, respectively. In image *f* the ^{52}Fe content of the stomach has decreased markedly, the duodenal loop is no longer visualized, but the jejunal area is well outlined. In image *g* virtually no ^{52}Fe is visualized in the stomach and duodenum, but the jejunal area continues to be well outlined. The ^{52}Fe can be seen in two places—the ascending colon and the jejunum—in image *h*, and it is noted that the rate of absorption has decreased considerably. In image *i* further movement of ^{52}Fe into the colon is seen, the jejunal area is still well outlined, and the absorption rate is small but still detectable.

The above results suggest that the delayed phase of iron absorption in man is related to the iron retained in the proximal small bowel. Fawwaz et al. hypothesized that this technique would be equally applicable to study of the intestinal site of absorption of virtually any appropriately labeled substance. They also show that "the use of the scintillation counter in its various forms has made possible the investigation of a large number of problems that would otherwise have been extremely tedious if not impossible."[21]

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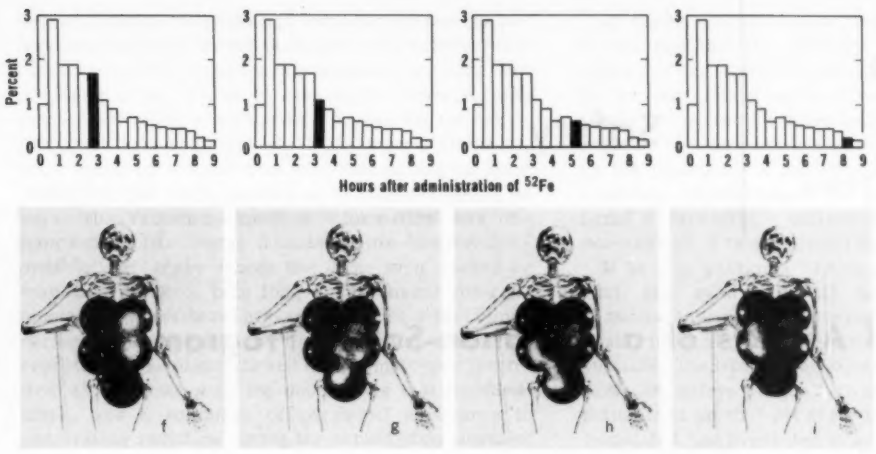

Fig. 8 Abdominal photoscans in a normal subject and plot of the percentage of ^{52}Fe absorbed at the time.

(a) 0 to 30 min
(b) 30 min to 1 hr
(c) 1 to 1.5 hr
(d) 1.5 to 2 hr
(e) 2 to 2.5 hr
(f) 2.5 to 3 hr
(g) 3 to 3.5 hr
(h) 5 to 5.5 hr
(i) 8 to 8.5 hr

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Further Selected Reading

Since no attempt was made to present an in-depth discussion in some areas—sensitivity, resolution, etc.—the following, although not cited, are presented as suggested readings.

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STATIC ELECTRICITY IN ANESTHETIC FLOWMETERS ELIMINATED BY RADIOACTIVE PISTOL

J. Hagelsten and Ole Larsen,
Brit. J. Anaesth., 37: 799-800 (1965)

An instrument containing 0.1 mc of ^{137}Cs in a protective covering mounted on a handle has been developed in Denmark to eliminate static electricity in anesthetic flowmeters. The accumulation of this charge causes the flowmeter bobbin to stick and may introduce a 35% error. Movement of the eliminator around the sticking bobbin neutralizes the charge and allows free rotation in seconds, a process that requires 4 to 8 hr under natural conditions. When not in use, the eliminator is kept in a lead box. The apparatus has been approved by the Danish Ministry of Health. (Martha Gerrard)

Section V

Isotopes and Radiation Technology

Safety

Legal Aspects of a Radiation-Safety Program*

By J. Lloyd Smith

The purpose of this article is to acquaint the health physicist in particular, and workers in the field in general, with some of the legal benefits of making and keeping good radiation-exposure records. Three lawsuits are referenced and discussed in which such records were invaluable. There is some discussion concerning the information that should be recorded, the type records that are admissible in a court action, and how long radiation records should be kept. Some conclusions are drawn as to why the general public is so fearful and confused regarding all radiation, and attention is called to the obvious need for continuing good records.

The radiation-safety program of an industrial concern or laboratory should have as its primary purpose the protection and safety education of its employees, individually and collectively. Further, the program, although not established for the purpose of providing evidence in an injury claim or a lawsuit, should also provide adequate records that can be used if a radiation injury or damage claim is made or a lawsuit is filed.

It is not the purpose in maintaining the records to prevent an employee, former employee, or even a third party from collecting on a rightful claim. In fact, after a claim of radiation injury is made and the necessary investigation reveals that the claim has merit, a settlement should be forthcoming. However, if it appeared evident from the investigation that the claim was without merit, the party against whom the claim was asserted would, of course, deny the claim. If the person alleging injury or damage filed a lawsuit, it would then be up to the courts to make a determination from the facts presented by both parties. Good radiation-exposure records are vital to the proper defense of such a lawsuit. In fact, a basic aspect of the whole defense structure of the suit would probably be the radiation records.

The need for good radiation-exposure records was exemplified in the recent trial of three radiation lawsuits. The suits brought by or in the name of former employees of a government contractor were the first of their kind and were tried concurrently. They were brought under the Federal Tort Claims Act in a Federal District Court. In each of the three suits, exposure to radioactive and toxic substances was alleged, and it was further alleged that, as a result of the exposure, one person (plaintiff *A*) was suffering from chronic lymphatic leukemia; the second person (plaintiff *B*) suffered from malignant lymphoma of the Hodgkin's disease type, with death occurring after the suit was filed; and the third person (plaintiff *C*) had suffered and died from acute granulocytic leukemia.

There were numerous allegations made by the plaintiffs in these suits. For instance, it was asserted that the U. S. government failed to properly supervise the "health and safety program," failed to require adequate training programs, and failed to properly supervise the contractor who was dealing with a very dangerous substance. The latter assertion was an attempt to establish nuclear material as an inherently dangerous instrumentality and hence to show that the government was engaged in an ultrahazardous activity. It was further alleged by the plaintiffs that the contractor failed to provide adequate safeguards, failed to furnish a safe place to work, failed to provide adequate medical supervision, and failed to provide proper equipment and protective clothing, with particular emphasis on the respirators or gas masks needed to screen out any toxic gases or radioactive materials. There was considerable controversy about the white puff or cloud that results when uranium hexafluoride is released to the atmosphere. It was also claimed that, as a result of these alleged failures, exposures to radioactive and toxic gases occurred and the above-mentioned conditions or diseases resulted.

After a review of the records, it was concluded that the claims were without merit; however, as can readily be understood, a considerable amount of work

*Reprinted, with permission, from *Nuclear Safety*, 7(1): 6-11, 18 (Fall 1965).

was required to properly and formally answer the allegations in court. Those in charge of the contractor's radiation-safety program accumulated records and prepared a breakdown of each of the three former employees' work assignments by year for the entire period of their employment (15 years for plaintiff *B*), their duties in the different jobs held during the time employed, and their exposures to radiation fields (if any). The radiation-exposure information was obtained from film-badge dosimetry, air-monitor records in the areas where the three men worked or may have worked, both long-term general environmental and short-term air-sampling records obtained during health-physics surveys, complete urinalysis reports on the plaintiffs and those employees identified as coworkers in the depositions that had been taken, and a summary of personnel exposures to penetrating radiation during the period of employment of the plaintiffs and others identified in the development of the proof.

In order to have the opinion of an independent and impartial expert, the accumulated information was sent to Dr. *D*, a health physicist and Professor, Department of Radiation Biology, School of Medicine and Dentistry, University of Rochester. After examining the information and making calculations, Dr. *D* reached certain conclusions, and his calculations and conclusions were then sent to two eminent physicians, Drs. *E** and *F*,† for their review and opinion.

At the trial of the cases, Drs. *D*, *E*, and *F* testified to the fact that the plaintiffs *had not* contracted or developed the diseases complained of as a result of the exposures they had received. Such testimony with respect to certain of these claims would not have been possible had the radiation-exposure material provided by the contractor's radiation-safety group been unavailable. Furthermore, the government would have found it exceedingly difficult to defend these actions successfully if this material had not been available.

It is also pertinent to note that the courts, in general, are unfamiliar with the subject of radiation, radiation injury, and the terms and usage common to health physicists and physicians in discussing these subjects. Therefore it behooves those in charge of the radiation-safety program to see that the records are maintained so that all needed information can be accumulated and presented in a clear and concise manner and in layman's terms whenever possible.

The fact that the evidence in the above-referenced case was well prepared by the contractor and equally well presented by the government is shown in the Court's Memorandum supporting the opinion and final order in the cases. The following excerpts from the Memorandum‡ are quite long; however, they are considered important to show the type of evidence that was presented, the Court's understanding of the evidence, the Court's understanding of the facts involved, and the conclusion reached:

> Plaintiffs contend that the amount of the dose is unimportant in evaluating injuries caused by radiation. This contention was supported by the testimony of Dr. *G*, who was of the opinion that dosage could only affect an individual according to his own susceptibility and vulnerability. Doctor *G* could not give any report that correlated Hodgkin's disease with radiation; nor could he establish permissible levels for human exposure to radiation. He stated that a human being's susceptibility or vulnerability to radiation could not be measured. He did not know the amount of the dose that any of the plaintiffs received throughout their work period for the *Contractor*. He was of the opinion that exposure of 2 rads of alpha radiation over a ten year period could cause leukemia.
>
> Doctor *H*, who like Dr. *G*, testified as plaintiffs' witness, disagreed with Dr. *G* on the question of whether the amount of dose of radiation was important in evaluating injuries caused to human beings. He was of the opinion that the amount of the dose was important. He knew of no specific worker having been injured while working within the permissible limits fixed by the I.C.R.P.[§] and the N.C.R.P.[¶] He did not know of any specific study groups on radiation that correlated chronic lymphatic leukemia or Hodgkin's disease with radiation exposure.
>
> Doctor *E* stated that there was no evidence from the Japanese tests and the Spondolytic English tests that a dose under 100 rads caused leukemia. He was of the opinion that the exposures of plaintiffs to radiation were not sufficient to cause either chronic or acute lymphatic leukemia or acute or chronic granulocytic leukemia, or

*Dr. *E* is a graduate of Tufts University Medical School. He is Clinical Professor of Medicine, Tufts University, Director of the Laboratories at Boston City Hospital, Chief of the Third Tufts Medical Service at the City Hospital, Fellow of the American College of Physicians, and Diplomat of the Board of Internal Medicine. He is a member of the American Medical Association, the Massachusetts Medical Association, and a number of scientific societies, including the International Society for Hematology, European Society for Hematology, American Society for Hematology, the Cancer Research Society, and the Society for Experimental Biology in Medicine. He is Director and a member of the Executive Committee for the Massachusetts Division of the American Cancer Society. He was Deputy Director of Research from 1952 to 1954, in Japan, of the Atomic Bomb Casualty Commission, and he investigated the blood of Marshallese who were irradiated on Mar. 1, 1954.

†Dr. *F* is a medical graduate from Oxford University, England. He also studied medicine at Balliol College and returned to Oxford for further degree work. He is now associated with Memorial Hospital in New York City, James Ewing Hospital in New York City, Bellevue Hospital in New York City, and Sloan-Kettering Institute for Cancer Research in New York City. He is a member of the National Academy of Sciences Committee concerned with the effects of ionizing radiation on the blood and consultant to the Office of the Undersecretaries of Special Political Affairs at the United Nations (UN) for the Scientific Committee on the Effects of Atomic Radiation. He wrote portions of UN reports on effects of radiation on the individual. He is on the subcommittee of the National Academy of Sciences concerned with the acute radiation injury in man, and he is a member of the American Medical Association, the British Medical Association, the American Association for Clinical Investigation, the American Society for Experimental Pathology, and the Biochemical Society. He is also a Diplomat of the American Board of Pathology.

‡References, in the quotations from the Court's Memorandum, to the names of the parties, witnesses, and locations have been changed and italicized, except for reference to the U. S. government.

§International Commission on Radiological Protection.

¶National Council on Radiation Protection.

Hodgkin's disease. He stated that radiation did not aggravate leukemia.

Doctor *F* was of the opinion that the doses of radiation to which plaintiffs were exposed were not sufficient to cause any of plaintiffs' disabilities. He stated that if the so-called linear theory should be adopted in the *Plaintiff C* case that the odds were 24 to 1 that *Plaintiff C* contacted the acute granulocytic leukemia from natural causes rather than from radiation. He was of the opinion that radiation did not cause *Plaintiff C*'s disability and death. Neither Dr. *E* nor Dr. *F* believed in the linear theory, i.e., that small amounts of radiation repeated over a period of years could cause leukemia and Hodgkin's disease.

Doctor *D* calculated the integrated radiation dose of absorbed uranium to each plaintiff. His report shows that *Plaintiff A*'s potential exposure was to a soluble form of uranium. In the *Contractor's* normal operation, the system is sealed and the activity in the air is not above normal background levels. But, in the event of cell changes, if the system is not completely purged, small amounts of residual UF_6, which is a soluble form of uranium, may hydrolyze and be released as a puff of white smoke in the air. The recommendations of the N.C.R.P. and the I.C.R.P. for maximum permissible air levels of soluble forms of natural uranium are set so as to provide a safe margin between industrial practice and a level which would produce kidney injury caused by the chemical toxicity of uranium. *Plaintiff A*'s exposure was to natural and slightly enriched uranium. Doctor *D* was furnished data on *Plaintiff A*'s urinary samples and his fellow workers over many years, including measurements of the uranium content and the number of alpha disintegrations per minute on each sample. This permitted a calculation as to the specific activity of the material to which the workers were habitually exposed. Doctor *D* was of the opinion that if *Plaintiff A* and his co-workers sustained any injury from radiation particles it would be to the kidneys. If kidney damage did not occur, he was of the view that absorbed uranium would be only a fraction of the radiation levels adjudged safe by the national and international experts. The medical reports of the plant showed that *Plaintiff A*'s kidneys functioned normally.

Doctor *D* made his tests to determine whether or not *Plaintiff A*'s work environment and that of his fellow workers carried with it an appreciable risk of overexposure from absorbed uranium. He examined the urinary measurements of 61 workers who were employed in the same sort of work and the same plant environment as *Plaintiff A*. He dealt with measurements of 1548 urinary samples collected for the ten year period from 1950 through 1959. The radiation risk over the period of *Plaintiff A*'s employment was evaluated by using the average of the many urine samples provided by him and his co-workers. The average total dose to each worker in *Plaintiff A*'s category over the ten year period is equal to 2.2 rems to the bone. Dr. *D* felt that the dose to the bone marrow would generally be estimated at one-half to two-thirds of this amount. His report shows that if *Plaintiff A* had lived in parts of Illinois where the radium in drinking water is somewhat higher than at *XYZ*, his yearly dose from natural sources would have been 0.2 rem per year and his 10-year dose from natural sources would equal 2.0 rems. The report also shows that the average per capita dose from diagnostic x-ray in the United States is 0.17 rem per year, or equal to 1.70 rems for a ten-year period.

The average count on 32 urine samples supplied by *Plaintiff A* himself is 0.2 alpha counts per minute per 100 ml urine. This amounted to a yearly dose of 0.05 rem or 0.5 rem over a period of ten years. Doctor *D* was of the opinion that *Plaintiff A* was more careful than the average workman and he consequently absorbed less uranium.

The Dr. *D* report shows that *Plaintiff C* was exposed to a soluble uranium compound. His potential exposure was to uranium hexafluoride, which is soluble in body fluids. When inhaled from the air it goes into the lungs and passes rapidly into the blood stream. About two-thirds of the amount inhaled is excreted in the urine within 24 hours. The remainder is temporarily taken into the skeleton and the kidney. Other soft tissues contain very small amounts. The organs which receive the greatest radioactive dose from the soluble compound are the skeleton and the kidney.

Doctor *D*'s report shows that *Plaintiff C* worked under conditions similar to *Plaintiff A* . . . .

Radiation experts are of the opinion that functional disability will not be caused from a dose of radiation not exceeding 0.3 rem per week for an entire lifetime. The tests show that the radiation dose received by *Plaintiff C* during his period of work was negligible. His dose was approximately one-sixtieth of 0.3 rem per week or 15 rems per year, which, as previously indicated, is the permissible dose for occupational exposure set by the I.C.R.P., N.C.R.P., and National Research Council.

Doctor *D*'s report shows that *Plaintiff B* was exposed to soluble uranium compounds and that airborne uranium compounds will accumulate in the lungs and the lungs become the organ where damage will occur. One day after the inhalation of soluble uranium compounds, the amount remaining in the lung is negligible. The uranium leaving the lungs enters the blood stream, and about two-thirds of this quantity is excreted through the kidneys within twenty-four hours. The remainder is temporarily taken into the skeleton and the kidney. Other soft tissues contain very small amounts; hence the skeleton and the kidney are the organs which received the largest dose of soluble compound. *Plaintiff B* was sent to the dispensary for checks on six occasions when a possibility of exposure existed. He was exposed to a mixture of natural and enriched uranium. If any injury occurred from absorbed natural uranium, it would be expected to occur to the kidney. If his kidneys were not injured, the reasonable assumption is that the radiation due to absorbed uranium would be only a fraction of that adjudged safe by national and international experts.

The report discloses further that the chronic and acute exposures of *Plaintiff B* over his fifteen year period of work service amounted to a total of about 4 rems. This was well within the range of radiation which is sustained by the non-industrially exposed population from natural sources in different parts of the world. It was not a contributing factor in the opinion of Dr. *D* for *Plaintiff B*'s fatal disease . . . .

The Court recognizes Dr *G*'s qualifications as a specialist in internal medicine. In weighing his testimony, however, consideration must be given to the fact that he has not done research or made special studies in the field of radiation. Nor has he made special studies of industrial workers who, at times in the performance of their work, are exposed to varying amounts of radiation.

That plaintiffs were not exposed to a sufficient amount of radiation to do bodily harm is established by the testimony of two eminent scientists, men who are working on the frontiers of knowledge, men who have done research and made special studies in radiation, men whose intellectual integrity was patent and who served only the cause of truth and not a partisan viewpoint.

The Court is of the opinion and holds that plaintiffs failed to establish a causal relation between their work for the *Contractor* and their resulting diseases or injuries by a preponderance of the evidence. The Court is further of the opinion and finds that plaintiffs failed to show by a preponderance of the evidence that the Government was guilty of any act of negligence that proximately caused their diseases or injuries. The Court, therefore, concludes that plaintiffs have failed to establish a case of liability against the Government under the Federal Tort Claims Act . . . .

It may be noted from the above quotations that the court was very much aware of the value of the radiation-exposure records and the calculations made by Dr. *D*. It is also evident that the testimony of Drs. *E* and *F* was dependent on those records and the calculations made by Dr. *D*, which were based on the maximum possible exposures, rather than the minimum exposures. At this point I would like to emphasize that it is important to be sure that the information provided to the health-physics expert provides him with the maximum exposures to which the complainant or plaintiff and his coworkers could have been exposed, rather than the minimum amount. Also, it should be made clear to the experts that this is the case. They will then be able to provide positive statements or opinions rather than weak equivocations. The findings of the court tend to bear out the assertions made by Edward Forgotson, M.D., L.L.B.,[1] when he said:

> Health physics testimony and evidence is so important because it protects medical experts by assuring that their testimony and opinions are predicated on the worst possible as well as the most probable exposure. When the medical diagnosis, and the purely medical basis for it, coupled with the maximum credible and the most probable dose of radiation received, is established, the probability of a given disease being produced or aggravated by the exposure of radiation can be stated with a high degree of certainty. It must be noted that on the basis of medical findings (physical and laboratory) alone radiation can be ruled out in many cases.
>
> The most important thing that the health physicist can do is to have his records complete. The records protect the health physicist in the same manner as the health physics testimony and data protect the medical experts. In order to answer a hypothetical question, the health physicist is going to have to be given the facts that were involved in the alleged exposure. In order to do this, these facts should be substantiated by the records so that he will be protected on cross-examination which can be quite a vicious thing and in which, by and large, no holds are barred.

The question may now be asked: what records should be kept? Obviously, a detailing of the exact records that should be kept cannot be given without a thorough knowledge of the information that may be available to any particular group. There are, however, some records that every company or organization dealing with nuclear or radioactive materials should keep. For example, the data taken from the film badges should be well documented and retained, as should air-monitoring records, whether of long term, of short term, or for a special job. Further, these records should be properly authenticated. Work histories should be kept, preferably in the form of a daily work log. Special investigative reports should be kept on all releases or incidents involving nuclear or radioactive materials. Also, it would be good to keep at least one copy of all plant manuals that may be issued dealing with health and safety in order to show the practice, requirements, limits, etc., for any period of time. And, of course, it goes without saying that all medical records, urinalysis reports, X rays, blood-test reports, and the like should be kept. In short, each work group should keep those records which will provide the information needed to properly carry out their assigned work and keep those records in such a way as to provide a true picture of the facts and circumstances involved.

Another question that should be considered is: What records may be introduced into a court action as evidence? Before answering that question, however, let us define *evidence* and some companion words and terms. *Evidence* may be defined as any species of proof legally presented at the trial of an issue, by *oral testimony* or *documentary evidence*, to enable the court or jury to decide upon the question in dispute. *Oral testimony* is that given under oath by a witness in open court based on facts of which he has firsthand or personal knowledge. *Documentary evidence* may be defined as tangible objects (such as handwritten, typed, bound, or unbound documents; pictures; graphs; and maps) that are capable of expressing a fact or tend to establish the truth or untruth of matters at issue and are not objectionable under the rules of evidence. A document is itself the best evidence of its existence.

Evidence admissible in a court action is limited to that which is germane (i.e., relevant, material, and competent) to the issue involved in the action. The federal courts and many of the state courts permit the introduction, as evidence, of writings made as a memorandum or record of any act, transaction, occurrence, or event if made in the regular course of business and if it was the regular course of business to make such records. Note the all-important "ifs." The individual who made the records may not be available due to death or otherwise, or the records may have resulted from the work of several people, and therefore the records may be introduced by the custodian of the records, who may or may not have firsthand knowledge of the contents. For example, the Medical Director may introduce in evidence any of the medical records in his custody.

Whenever records are discussed, the question of time must also be considered. How long must the records be retained? Records dealing with radiation exposures, concentrations, or the lack thereof in laboratory or plant areas should be placed on a permanent-retention status because the field of radiation damage or injury is so new to the courts and the time period in which such injury or damage may evidence itself is so indefinite that no one can say, at this time, how long it will be necessary to keep such records.

There seems to be little doubt that those who deal with radioactive materials will continue to be faced with claims alleging radiation injury or damage, probably in increasing numbers, at least for some extended time. Many, if not most, of these claims will result from fear or ignorance of the facts concerning

radiation, rather than from actual radiation injury or damage. The natural incidence of the disease or condition from which the claimant suffers is many times disregarded by both the claimant and his attending physician once it is determined that the ailment can or could possibly be the result of radiation and the claimant has been, in any manner, connected or associated with radiation or with a concern dealing in or with radiation products.

Such an attitude is understandable if we remember that nuclear energy and the resulting effects were introduced to the public by the bombing of Hiroshima and Nagasaki. The public associates nuclear energy and radiation with death and destruction, and too little thought is given generally to its many lifesaving and industrial uses. It has been said that, if gasoline had been introduced to the public as the napalm or incendiary bomb instead of as fuel for the internal-combustion engine, there would probably be an entirely different view taken of its use and refining processes. We also have continuous reminders by the civil-defense groups of the extreme hazard of radiation. They are, of course, considering fallout from bombs that may deliver doses in the hundreds or thousands of rads. No one will disagree that such conditions would present an extreme hazard; however, if we are to remove the unwarranted anxieties of those employed in or about radioactive materials or sources, the public must be educated to the differences between the civil-defense types of radiation hazard and the hazards encountered in industrial plants and laboratories. By "differences" we do not imply that the radioactive materials found in industry and in the laboratory are not harmful *if mishandled*, but we do mean that with proper precautions such materials can be handled safely. When this idea has been properly presented and accepted, claims based on unreasonable fear or ignorance should decline rapidly. However, it will continue to be necessary to keep records, since there will always be those who will become careless and get hurt and, unfortunately, some who would rather sue than work. Additional information on radiation-injury litigation is contained in Refs. 2 to 5.

References

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Section

VI

Isotopes and Radiation Technology

Isotopes Around the World

^{60}Co Irradiation Facility of the L. Ya. Karpov Physical Chemistry Research Institute*

Abstract: The 200-kilocurie irradiation facility of the L. Ya. Karpov Physical Chemistry Research Institute is described.

The KP-140 irradiator, designed and fabricated at the L. Ya. Karpov Physical Chemistry Research Institute, Moscow, contains 200 kilocuries of ^{60}Co. It was put into operation in 1964. A special feature of the facility is the use of compressed air both to cool the source and to move it to and from the storage and irradiation positions. The source-moving equipment is located outside the irradiation facility; thus the operating space is not cluttered.

The source is made up of 81- by 11-mm-diameter ^{60}Co pellets, each individually contained in a stainless-steel capsule with a self-locking threaded cover. When not in use, the capsules are stored in 20 U tubes, seven capsules to a tube. The lower portions of the tubes are sealed into a leaktight storage cell that forms the lower part of the irradiation facility (Fig. 1). The space around the tubes is filled with iron shot.

One arm of each U tube is connected outside the cell to a supply of compressed air, and the other is connected to an 80-cm-high tube in the irradiation portion of the chamber. The 20 irradiator tubes are arranged on a circular base, thus forming a cylindrical array in which the object to be irradiated is placed. The pipes connecting the storage tube to the irradiation tubes are bent and can be rotated so as to change the cylinder-base diameter from 20 to 120 cm (Fig. 2) and thus vary the radiation field in the cylinder. The field strength is also dependent on the number of pellets moved into the irradiator tubes.

When the pellets are to be moved from one tube at a time, the air reservoir is pressurized to about 0.4 atm gage, the valve between the compressed-air

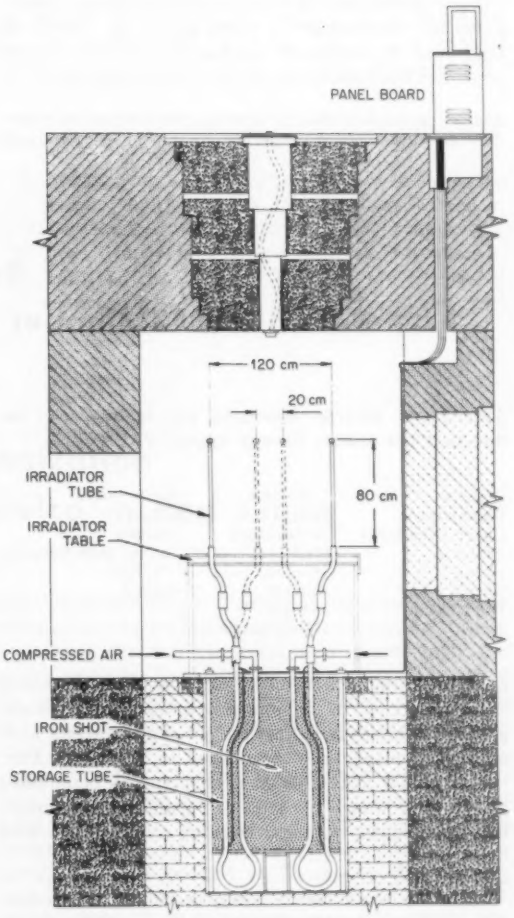

Fig. 1 The KP-140 ^{60}Co irradiation facility.

*Based on an article (in Russian) in *Atomnaya Energiya (USSR)*, 18: 546-548 (1965).

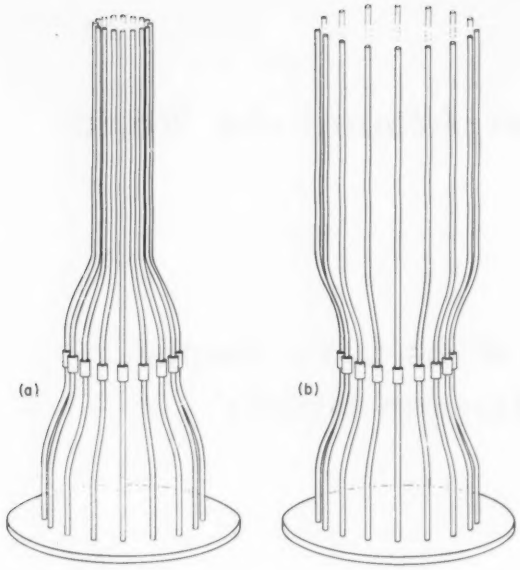

Fig. 2 Irradiator tubes (a) *arranged for smallest cylinder-base diameter and* (b) *opened up.*

tube and the source-storage tube is opened by remote control, and the air pushes against a piston in the storage tube which in turn pushes the source pellets into the irradiator tube. The operation is repeated until as many sources as are needed for the operation have been moved into place. The time required is 1 to 1.5 min. If several or all are to be moved simultaneously, a pressure of 1 atm gage is built up in the storage reservoir.

When the pellets are to be returned to storage, the valves may be closed one at a time; shutting off the compressed air supply drops all the pellets simultaneously. A braking action in the U tube prevents impact shock at the bottom.

The location of the pellets in the tubes is monitored by pneumatic sensors, which relay information to the control panel. A specially designed air lock helps prevent any unauthorized entry to the irradiation chamber.

In tests on the system under conditions corresponding to 15 to 20 years of actual operation, the ^{60}Co capsules remained pressuretight and showed negligible wear. After a year of operation, the equipment was still functioning dependably.

(Martha Gerrard)

Use of Radioisotopes in IAEA-Member Countries of Africa*

Twenty-one African countries are members of the International Atomic Energy Agency (IAEA):

Algeria	Liberia	South Africa
Cameroon	Libya	Sudan
Congo (Kinshasa)	Madagascar	Tunisia
Ethiopia	Mali	United Arab Republic (UAR)
Gabon	Morocco	Uganda
Ghana	Nigeria	
Ivory Coast	Senegal	
Kenya	Sierra Leone	

IAEA assists, in agreement with the governments of these countries, in implementing national development plans and in ensuring that the potential peaceful uses of atomic energy are considered in future planning.

A regional center for Arab countries is maintained in the UAR where persons are trained in radioisotope techniques; regional training courses have been held in Congo, Uganda, and the UAR; and a regional course for African participants in the repair and maintenance of nuclear instrumentation will be held, probably in Sierra Leone, in 1968. Since training in the more specialized aspects of atomic energy cannot yet be obtained in Africa, more than 200 Africans have studied overseas under the IAEA fellowship program.

Agriculture

Radioisotope-labeled fertilizers are being used in studies designed to increase the efficient use of fertilizer on rice in Madagascar and the UAR and on maize in the UAR; interest in the maize project has been expressed by Ghana. Nuclear techniques are being used in production studies on cocoa, peanuts, and rubber in Ghana; on groundnuts and sorghum in Senegal; and on olives in Tunisia. New wheat strains, developed with radiation, have recently been shown to be superior to local and common strains in Libya, Tunisia, and the UAR. The possibilities of eradicating the Mediterranean fruit fly by the radiation-

*Extracts from "Atomic Energy in Africa," an abbreviated version of a paper prepared for the 8th Session of the Economic Commission for Africa, *International Atomic Energy Agency Bulletin*, 9(2): 7-14 (1967).

sterilized-male technique are under extensive study in Tunisia and Morocco, and research on the tsetse fly is active in Uganda.

Portable neutron probes for soil-moisture determination, which are valuable in studies on conservation and use of water in crop production, have been in use for some time in Algeria, Ghana, Ivory Coast, Kenya, Madagascar, Morocco, Senegal, and the UAR.

Food Preservation

Although facilities for radiation preservation of food are not yet available in Africa, the need for such studies in the countries where the food is produced and where the process would be used is recognized. Consideration is being given to including irradiation in the broad research programs already under way in Mali, Niger, and other countries on conventional methods of preventing fish spoilage by insects. The major problem is finding an inexpensive insect-proof package to prevent reinfestation. Pilot studies being carried out in Europe on combining fish irradiation and refrigeration are the first phase of a program that will be continued in Abidjan. Likewise, tests are being made in Europe on irradiation of meat, bone, and blood meals as the first phase of a project to enable Chad to export such products. A limited research program to provide knowledge of the potential and limitations of food irradiation is under way in Nsukka (Nigeria) and Tunisia. A training course in this field will be held in cooperation with the United States Government in 1967.

Hydrology

Isotope techniques are proving valuable in Kenya, the UAR, and Tunisia in locating the sources of groundwater recharge and determining the direction and velocity of groundwater movement as an additional tool in efficient use of available water resources. These techniques have also been successful in determining the availability of groundwater for municipal purposes in Lusaka (Zambia).

Industry

Recent work in the UAR has clearly demonstrated the range of useful applications of radioisotopes in industry, tracer experiments having been made in oil fields, steel plants, the glass industry, fertilizer factories, and ore-dressing plants. Nondestructive testing of ingots by gamma radiography and highway subsoil moisture determination by radioisotopic methods were also introduced. Consideration is being given to the use of isotope techniques in industrial development schemes now being undertaken in Ghana.

Medicine

Radioisotope laboratories for diagnostic purposes are in operation in Algeria, Congo, Kenya, Morocco, Nigeria, Senegal, South Africa, Sudan, Tunisia, the UAR, and Uganda; and liver cancer, sickle-cell anemia, and various types of malnutrition are being actively studied. (Martha Gerrard)

New Reorganization of Peaceful Nuclear Service in Red China*

With the upheaval in Red China caused by the cultural revolution not yet over, in January 1967 Peking proceeded to an important reshuffling of personnel working in the atomic energy service (more so for military ends than for public objectives).

Factories intended for bomb manufacture are seriously disturbed in Sin-Kiang; they are being reorganized, but the details of this operation are scarcely noticeable outside. Apparently their direction is no longer assumed by Dr. Chien San Chieng. On the other hand, some information has been received, from as many Chinese sources as Japanese, and even from India, on the reorganization of centers for research or for isotope production—particularly the Shenyang center in Manchuria and the one in Lanchow for the production of enriched uranium. In principle at least, these are not answerable to military authorities.

An important change involves Mrs. To-Tsa Hui, wife of Dr. Chien San Chieng, who prior to the cultural revolution directed the Department of the Peaceful Atom; she has been replaced by Dr. Wou Lang, who was trained in China and thus is very "pro-Mao," whereas Mrs. Hui, coming from the Moscow Institutes, was considered to favor the Russians. Dr. Wou Lang is assisted currently by two professors, Bu Kuang Tung and Hou Wit Tou, who have

*Edited version of Li Yang, New Reorganization of Peaceful Nuclear Service in Popular China (in French), *Energie Nucleaire (Paris)*, 9(2): 137 (1967), translated, with permission, by Carol S. Gamble, Report ORNL-tr-1737.

replaced Hsiung Ching Lai and Chien Huang Tuan and are considered from their backgrounds to be pro-Russian.

Unaffected by the reshuffle are Chiang Cheng Lieh, Ting Yu, and Ko Mi Yang, specialists in nuclear applications in agriculture and medicine. Likewise, Dr. Wang Kan Chang is still department head for fundamental studies and research. In the meantime, a "diffuser of foreign sciences" has been added to the department; he is Fan Din Houa, who seems to have put a curb on the work of two Nobel Prize winners in physics under the pretext, no doubt, that the two did their research at Columbia University in the United States.

Interesting progress has been made in the fields of atomic energy and space.

Other changes are still to be made; yet research centers have continued to operate normally enough. On the other hand, it will not be the same for the military center of Sin-Kiang. (CSG)

THE WORLD OF RADIOISOTOPES, by J. N. Gregory
Published by Angus & Robertson Ltd., Sydney, Australia, 1966
(204 pp.; 52 plates, 22 line drawings)

Any comprehensive review of the uses of radioisotopes, which is presented both with scientific accuracy and in semitechnical language, is a welcome addition to the radioisotope literature. This book, written by the Chief of Isotope Division, Australian Atomic Energy Commission, is easy to read, but there is no talking down to the educated but nonscientifically trained reader. In the author's words, it is not intended to be "as a definition or authoritative textbook. It is simply a guidebook to a tour of the world of radioisotopes, which holds a lasting fascination in those people who work in it—a fascination which arises from a never-ceasing amazement at the basic simplicity and versatility of radioisotope methods." There is a good index but, unfortunately, no bibliography.

Both general and specific applications of radioisotopes are discussed, e.g., the use of radioisotopes as tracers for industrial process streams and their specific use in the mining industry. Whole chapters are devoted to radioisotopes in industry, in hydrology and civil engineering, in agriculture and the food industries, in medicine and medical research, and in the physical sciences. The first three chapters explain the fundamental principles on which the uses of radioisotopes are based, including detection and measurement methods. Throughout the book, ancillary equipment and processes are explained, for example, thermoelectric circuits for radioisotope generators, methods for introducing radioactive tracers into systems, and the carbon cycle in nature as applied to radioisotopic dating. A chapter on safety in handling radioisotopes includes a discussion of various safety principles, among which are shipping-container design and unsafe levels of contamination.

Specific data are quoted in many cases, for example, the cost of radiation-sterilized medical syringes, the production rate of ethyl bromide in the radiation-initiated process, specific results from a tracer-labeled sewage-stream test, and the exact copper content of coins analyzed by nondestructive radioisotopic methods.

The last chapter, "Conclusions and the Future," is an optimistic forecast of future uses of radioisotopes. (Martha Gerrard)

MANUAL OF RADIOISOTOPE PRODUCTION

The *Manual of Radioisotope Production* published by the International Atomic Energy Agency (IAEA) in Vienna in 1966 (Technical report No. 63) was compiled primarily for the benefit of small reactor establishments with a modest program of radioisotope production for local requirements. It is not intended as a comprehensive document but as a guide to solutions of problems that may be met in early stages of production. Numerous references are included.

Part I of the manual describes general aspects of radioisotope production programs—initial planning, facility design, waste management, handling of solutions for medical use, product quality control and assay, and radiation protection practices.

Part II gives detailed methods—both irradiation data and separation procedures—for the 15 radioisotopes in most common use: ^{82}Br, ^{58}Co, ^{51}Cr, ^{64}Cu, ^{18}F, ^{198}Au, ^{131}I, ^{59}Fe, ^{28}Mg, ^{24}Na, ^{32}P, ^{35}S, ^{90}Y, ^{42}K, and ^{65}Zn. The production methods were contributed by IAEA member states with long experience in radioisotope production: Canada, Nationalist China, Czechoslovakia, France, Hungary, India, Israel, Japan, Norway, Romania, Poland, Spain, United Kingdom, United States (both Oak Ridge and Brookhaven National Laboratories), and Yugoslavia. Several different procedures are given for each nuclide. Cobalt-58, for example, is made at six different installations, and each installation contributed a description of its techniques.

In addition, nuclear properties of these nuclides are listed, with no attempt to indicate best values, and typical uses are tabulated and referenced. (Martha Gerrard)

Section

VII

Isotopes and Radiation Technology

Miscellaneous

AEC Activities

Division of Isotopes Development Provides Summaries of Offsite Contracts

During FY 1967 the Division of Isotopes Development (DID) of the USAEC was involved in the sponsorship of approximately 100 offsite contracts in several major development areas: (1) Isotopes Production and Materials Development, (2) Thermal Applications, (3) Radioisotope Applications Development, (4) Radiation Processing Applications Development, and (5) Radiation Preservation of Foods. It is believed that the following brief summaries of these contracts will be of considerable interest to many of our readers, since they present an overall picture of the breadth and depth of the DID program. In each category the summaries are followed by a listing (in the same order as the summaries) of the principal investigators and addresses of the contractors.

This information will be updated periodically, as new contracts are administered or as scopes change.

Isotope Production and Materials Development

Isotope Device Safety Testing Program. AT(40-1)-3610, AT(11-1)-1409. Underwriters' Laboratories, Inc. This program will develop technical information and recommend test procedures for developing standards and performance test criteria applicable to devices that use radioisotopes to accomplish their intended functions. It will also provide information to the device manufacturers and regulatory agencies to assure public safety and confidence in devices employing radioisotopes. This program will include a detailed investigation and analysis of (1) normal usage and normal environment that may affect the devices and (2) abnormal usage and conditions that may result from accidents; exposure to fire, corrosive gases, or liquids; explosive vapors, etc., that may occur during the lifetime of the devices.

The objectives will be reached using the following techniques: (1) review product specifications and quality control procedures, (2) review environmental conditions of installation and use, (3) inspect representative installations, (4) review available accident records, (5) initiate a plan of investigation to identify and weigh the safety characteristics of the devices under investigation; (6) carry the tests and investigation of representative devices to such a point that sufficient data are available to indicate the performance of the test samples under the prescribed test conditions, and (7) review items 1 through 6 and prepare for delivery to the Commission a Safety Standard covering the class of device under investigation.

The devices studied include radiographic, gaging, teletherapy, and other selected isotope devices.

Representative gaging and portable radiographic devices have been tested, and test procedures (including flexing, compression, tension, and kinking) have been proposed. The results are under review.

INVESTIGATORS AND ADDRESSES

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Thermal Applications

Radioisotope Applications in Hydrospace. AT(04-3)368, Project No. 5. Aerojet-General Nucleonics. The objective of this program is the investigation of the applications of radioisotope thermal power in hydrospace. One potential requirement is for dynamic-conversion radioisotope power sources in the range from 2 to 20 kw(e). Conceptual design effort has been directed to (1) fuel specifications, (2) Dowtherm Rankine-cycle technology, (3) safety engineering and testing, (4) Dowtherm test loop, (5) mission/technology interface study, (6) alternative boiler conceptual design, and (7) handling and transport analyses.

Application of Radioisotopes to Manned Spacecraft and Life-Support Systems. AT(04-3)-575. AiResearch Mfg. Division, Garrett Corp. This work involved the study of potential applications of radioisotopes to the operation of spacecraft life-support systems. Planned

spacecraft missions and the life-support system processes required were considered. The integration of radioisotope thermal sources into the specific process functions was analyzed, and weight comparisons were made between isotope energy systems and systems operated by fuel cells and solar cells. Studies were made of radioisotope selection and source design considerations, and trade-offs were made between single-source and multiple-source systems. Detailed studies were made on the applications of isotopes to the processes of carbon dioxide removal, refrigeration, water and oxygen recovery, spacecraft heating, contaminant control, biological systems, and lunar material processing.

It has been determined that isotope heating shows considerable promise in extended spacecraft missions for processing water, regenerating the spacecraft atmosphere, heating the vehicle, and decomposing solid wastes. Radioisotopes are shown to be potentially suitable for providing refrigeration and for the processing of lunar materials, but proof of these applications awaits prototype construction and testing.

Design Study of Integrated Life-Support System for Aerospace Application Utilizing Radioisotopes for Thermal Energy. AT(04-3)-739. Lockheed Missiles & Space Co. The joint AEC/Air Force 9-month study will identify an integrated aerospace life-support system providing the highest reliable usable payload with the lowest possible launch weight, and determine the optimum energy source after considering radioisotopes for direct thermal energy. Feasibility evaluations and conceptual design of a system for a 180-day mission and a two-man crew will be performed. Design criteria for the life-support system and radioisotope thermal-energy (RITE) sources will be established. Candidate concepts for the life-support system functions, such as urine distillation, catalytic oxidation of trace contaminants, and Sabatier reactor for CO_2 reduction, will be evaluated and system heating requirements will be determined. A study of candidate radioisotopes, such as ^{60}Co, ^{90}Sr, ^{147}Pm, ^{210}Po, and ^{238}Pu, will be made to optimize the isotope selection in the area of minimum heat-source weight, minimum shield weight, size, and safety.

On the basis of the component evaluations of the life-support system and the heating requirements, several complete integrated systems will be compared, and the best integrated system maximizing the use of RITE will be recommended to the AEC. After selection is made, component configuration and design support analyses will be performed. Layout drawings and component detail drawings will be prepared to allow fabrication of a functional proof-of-principle component.

Implantable Power Source for Circulatory Support Systems. AT(30-1)-3857. Westinghouse Electric Corporation. The principal objective of this program is to define an implantable power source, using isotopic heat, for use with human circulatory support systems in terms of a conceptual design that offers promise of being developable into useful hardware. Conceptual studies will be carried out to provide an integrated dynamic power-source design based on a steam Rankine cycle. The design will consist of an encapsulated radioisotope heat source, a heat-to-mechanical-energy converter based on the Meyer rotary expansion engine (or modifications thereof), a heat-rejection exchanger using blood as the cooling medium, a control system to regulate the power output, and ancillary equipment. Further conceptual studies will be carried out to evaluate major problems involved in the successful development of this power-source design concept, to provide cost and time estimates, and to compare this design concept with other potential concepts.

Study of an Implantable Power Source for Circulatory Support Devices. AT(30-1)-3858. Thermo Electron Engineering Corp. The objective of this study is to develop a conceptual design and characterize the development problems for an implantable radioisotope-powered source for circulatory support devices. The power source consists of an encapsulated radioisotope heat source, a Rankine-cycle engine that dumps the reject heat into a blood-cooled heat exchanger, and a control system to control the power output. The power unit will be implanted in the abdominal cavity and linked to a blood pump in the chest. Miniature steam engines are attractive for this application because of their high practical efficiency and reliability. The conceptual design will be optimized with respect to thermodynamic efficiency, radiation level, weight, and volume. The study includes parametric analyses of all system components and an evaluation of the candidate radioisotopes — ^{238}Pu, ^{147}Pm, and ^{171}Tm. The Rankine-cycle engine will be compared with other thermodynamic cycles. Major problems to be encountered in the development of a workable engine from conceptual design will be identified. Gross resources in terms of manpower, materials, and time to carry the conceptual design to a workable prototype model will be estimated.

Conceptual Design Study of Implantable Power Sources for Circulatory Support Systems. AT(04-3)-368, Project No. 7. Aerojet-General Nucleonics. The program consists of a parametric analysis and conceptual design study of a radioisotope-powered, modified Stirling engine for use as an implanted power source for an artificial heart. The 7-month program consists of (1) conceptual design of the engine, including detailed design evaluations; (2) parametric analysis, including a significant analog-computer evaluation; (3) identification of major problems; (4)

comparison of conceptual design with other possible engines; and (5) estimate of resources required for the development of a prototype engine. The engine being studied differs from the standard Stirling engine; it does not incorporate a crankshaft (or crankshaft bearings), but a reversing piston is provided to control the action of a conventional displacer piston. The engine extracts energy from radioisotope-heated helium working fluid in a closed cycle and accumulates the energy in the form of gas pressure in a suitable gas reservoir. The accumulated gas pressure is used to pump a secondary fluid through an appropriate bellows and a pressure-reducing system, which, in turn, provide the direct pumping power for an artificial heart. The system is operated in a pulsed mode that can be triggered either by a physiological input or by a constant-rate timing device. The exhaust gas from the pumping chamber is routed through a heat exchanger that rejects waste heat to the blood.

The engine will deliver up to 7 watts of pumping power, will weigh approximately 2.5 kg, and will occupy approximately 2 liters. The maximum radioisotope source temperature will be approximately 1200°F, and the overall efficiency is estimated at 21.8% after 10 years. Biological radiation shielding will be provided to control the dose rate at the surface of the engine.

Implantable Power Source for Circulatory Support System. AT(45-1)-2112. Donald W. Douglas Laboratories. The purpose of this contract is to prepare a conceptual design of an implantable power source (radioisotope-powered engine) for circulatory support systems within the human body. The engine is to power either an assist or total-replacement heart pump. The device consists of an encapsulated radioisotope heat source, an air engine type of thermodynamic converter, a heat exchanger using blood as the cooling medium, and a control system to regulate the power output of the engine. The heat source will be ^{238}Pu, ^{147}Pm, or ^{171}Tm. The thermodynamic converter will be capable of providing 7 watts of mechanical power and capable of implantation and operation in any orientation. The major factors considered in the design will be minimization of waste heat, radiation exposure, weight, and volume and the evaluation of position sensitivity, engine lifetime, and reliability of the various engine components.

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Radioisotope Applications Development

AEROSPACE

Nucleonic Mass Metric Instrumentation of Propellants for Aircraft. AT(49-11)-2951. Wright-Patterson Air Force Base; General Nucleonics Corporation. Continuous measurement of fuel mass in a shallow wing fuel tank independent of flight attitude and tank geometry is to be achieved as a result of the fact that radiation absorption is a function of the fuel mass between a number of radiation sources and their detectors. The detector is lithium-drifted silicon ($2\ cm^2 \times 2.0$ mm) or cadmium telluride (CdTe); the radiation source is ^{85}Kr, ^{60}Co, or ^{137}Cs. Computer analysis will be used to determine the number and placement of detectors and radioactive sources on the fuel tank at any geometry with wing deflection and wing twist being taken into account. Combinations of pitch and roll will be included in the attitude study. A system of minimum size, weight, and power is to be designed with a mass fuel-measurement accuracy of ±0.5% at full capacity.

Radiotracer Propellant-Gage Technology Program. AT(04-3)-539. Space Technology Laboratories, Inc. This program will investigate, evaluate, and advance nuclear measurement technology for application to spacecraft propellant gaging. A gaging-system improvement, made by replacing pressure–volume–temperature type density measurement with a direct density measurement utilizing a beta-emitting radionuclide and detector, will be evaluated. The goal is to develop a Density–Volume Propellant Gaging System that is accurate to 0.1% in the environment and configuration of a typical pressurant storage tank. Since propellants diffuse through all of the currently used flexible expulsion bladders, development of protective covers or coatings for the propellant gage detector is necessary. Teflon–metal laminates, silicone–rubber–nickel laminates, and Parylene C will be evaluated as detector coatings to withstand saturated N_2O_4 vapor for 1000 hr at 120°F. Replacement of ^{85}Kr with ^{39}Ar or some other pure beta-

emitting gaseous radionuclide in a radiotracer propellant gage will be investigated.

Isotopic Altimeter-Setting Indicator System—Digital Readout. AT(49-11)-2811. Systems Research and Development Service, Federal Aviation Agency; Panametrics, Inc. This research entails the development of an altimeter-setting indicator system using an isotopic source to measure atmospheric-pressure changes, which are converted to the corresponding altimeter-setting indication and digitally displayed. The system will operate at all elevations between 100 ft below, and 6600 ft above, mean sea level, giving readings to the nearest 0.01 in. with accuracy ±0.005 in. in the range 28 to 31 in. Hg. Up to 10 remote displays can be accommodated. The radioactive source and detector will have an effective life of at least 5 years, be safe to handle with normal precautions, and meet public safety criteria. The system will operate at temperatures between 0 to 60°C and withstand temperatures to −20°C without damage. Design emphasis will be placed on reliability and ease of maintenance.

Military Helicopter Formation-Keeping System. AT(11-1)-1471. Industrial Nucleonics Corporation. The objective of this research is the development of an operational relative-position-indicator system capable of simultaneously identifying and resolving the position of three other aircraft. The system will have a range accuracy of 10% from 50 to 500 ft and maintain comparable bearing accuracy with helicopter tilt of ±30°. Weight limits for the source and detector are 10 and 30 lb, respectively. Both a rotational scanning detector system and a stationary detector system have been considered, with research being focused on a stationary NaI detector system and the use of modulated radionuclide soft-gamma or X-ray sources as the most promising. As sources, ^{241}Am, ^{137}Cs, ^{60}Co, beta-excited X rays, and conventional X rays have been considered. A cathode-ray tube provides visual information display. Design feasibility has been demonstrated in ground tests; airborne tests will be conducted using a modulated ^{85}Kr-excited X-ray source.

CRIMINALISTICS

Development of Neutron Activation Analysis for Regulator and Criminalistics Problems. AT(49-11)-2321. Internal Revenue Service, Alcohol and Tobacco Tax Division Laboratory. Work on this project will include study of (1) neutron activation analysis for identification of narcotics and opiates and for determination of the geographic area of origin and international movement of illicit narcotics; (2) characterization of pharmaceutically pure drugs, with the objective of correlating the trace-element composition of the drug with manufacturing processes and other phases of preparation, control samples from the manufacturers being used to establish standards; (3) evaluation of the effectiveness of neutron activation for tracing the origin of raw materials such as grain, yeast, and urea and for linking a specific still with seized liquor; and (4) activation analysis as a means of detecting altered documents and counterfeit currency.

Neutron Activation Analysis for Scientific Crime Detection, AT(04-3)-167, Project No. 1. General Atomic Division, General Dynamics Corp. It is proposed to increase the effort in forensic applications of activation analysis to include (1) examination of ~100 samples of commercially available paints, papers, and glasses to ascertain their trace-element content; (2) study of hand-blank neutron activation analysis measurements of antimony and barium from gun firings; (3) modest studies of trace-element content of other evidential material such as rubber, narcotics, and bullets; and (4) investigation of stable-element labeling of various materials, e.g., bullets and leads, for trace-element content. Both neutron and photon activation would be used, with the induced radioactivity measured by NaI(Tl) or Ge(Li) spectrometry. The ultimate objective is use of such data and/or techniques for law-enforcement purposes.

Radioisotope-Excited X-Ray Fluorescence Analysis for National Security Needs. AT(40-1)-3561. Panametrics, Inc. The contractor shall explore and evaluate the potential application of radioisotope-excited X-ray fluorescence techniques to national security problems and to other problems related to the common welfare. The tracer techniques associated with X-ray fluorescence can be applied to many problems such as (1) theft prevention or tracing of stolen items; (2) monitoring stack gases and industrial wastes to prevent pollution; (3) prevention or identification of counterfeiting of items of value, documents, and bank-note paper; and (4) aiding of mail handling by methods of sorting or stamp canceling.

ENVIRONMENTAL

Radioisotope Water-Flow Measurement in High-Head Turbines and Pumps. AT(49-11)-2758. Bureau of Reclamation. A program jointly sponsored by the Department of the Interior and the Atomic Energy Commission is designed to examine the problems of water flow in turbines. Radioisotope methods and instrumented systems developed by the Denver Office, Bureau of Reclamation, Department of the Interior, for open-channel water-flow measurements are adaptable, in part, to the problem of accurately determining the rate of water flow through high-head turbines and pumps—a measurement that cannot be made as accurately by conventional methods. Research is being conducted to determine the minimum mixing length required to obtain adequate mixing. Techniques and equipment for injection of radioisotopes are being developed. A program of field testing is being formulated to test methods and equipment being developed.

Development of a Low-Level Radioactive Gas Tracer System for Natural-Gas Operations. AT(49-11)-2757. U. S. Department of the Interior. The aim of this research is to develop a system capable of unattended operation under field conditions for the use of very low levels of radioactive gas as a tracer in natural-gas production and storage. The basic detection element of the system is a methane-gas proportional counter using tritiated methane as a tracer. The detection system will allow quantitative measurement down to 5×10^{-15} curies/cm^3 with 90% confidence in 2 hr of counting, with anticoincidence techniques used to achieve this sensitivity.

The field application of the system demands rugged modular construction, with maximum weight of any single unit <70 lb and total length and girth within 100 in. The system will operate at atmospheric pressure, ambient temperatures 35 to 110°F, and relative humidity 10 to 95%.

A necessary part of this research will be the development of a device for injecting radioactively tagged gas into a gas-flow stream proportionately so as to maintain a preestablished tag level in spite of changes in flow rate, density, temperature, or other parameters. Control of injection level will be maintained through measurement of the tag level and regulation of the injection device through a servo system.

Methane Ionization Detector. AT(30-1)-3717. Melpar, Inc. Previous work has resulted in the construction and testing of a Methane Ionization Detector for use in coal mines. The device employs a simple gas-ionization detector that contains tritium as the ionizing source. The detector has been tested under the humidity and pressure conditions encountered in a coal mine. In addition, it has been subjected to the simulated vibrational conditions associated with a coal-mining machine. The present work is aimed at further testing on the cutting head of a mining machine. The end product is expected to be a monitor that is stable and reliable when mounted directly on a mining machine during the rigors of routine daily operation.

Mineral Exploration Systems Development. AT(49-11)-2784. U. S. Geological Survey. The purpose of this project is to develop mineral exploration systems based on nuclear methods of analysis. The first phase deals with a system for the measurement of silver in low-grade ores. Neutron activation of the silver by means of a portable neutron generator, and gamma detection equipment to count, discriminate, and time the decay rate of the activation spectrums will be used. All the operating characteristics of the system are to be tested and quantitated under simulated field conditions.

Nuclear Techniques in Soils Hydrology, Snow Gaging, and Related Applications. AT(49-11)-2773. U. S. Department of Agriculture, Forest Service. Previous work has been in the hydrology of soils, construction and use of a snow-density gage, use of water by plants, and tracing of hexadecanol in the investigation of retarding evaporation of water.

Four parts, all of which will use isotopes, are included in the current study. Part 1 involves the determination of the effect of aspect and cover on snow accumulation, metamorphosis, and melt. The objective of this study is to determine the mathematical relationship between the snow condition determined by the snow gage at the Central Sierra Snow Laboratory and conditions existing at various locations within 5 miles of the laboratory; hopefully, the centrally located snow gage can be used to indicate snow conditions at any other point within this area. Part 2 involves collection and analysis of soil samples. This work will continue since confirmation of soil hydrological properties by analysis is essential to the study of tracing water with isotopes. Part 3 comprises a study of soil water movement under saturated and nonsaturated soil moisture conditions. In a joint effort with Hazleton-Nuclear Science Corporation [contract AT(04-3)-676], attempts will be made to measure the movement rate and amounts of water in the soils studied, to determine the path and rate of soil water moving down a slope from hilltop to stream, and to determine water use by vegetation growing on a hillside. Part 4 involves the determination of water movement in plants, using isotope and heat-pulse techniques. Relationships between velocity of water movement in the stems and environmental driving factors, such as solar radiation, soil moisture availability, and temperature, will be determined in various types of vegetation indigenous to Sierra Nevada.

Snow-Water Conservation Studies. AT(04-3)-676. Hazleton-Nuclear Science Corporation. Work in progress is on (1) methodology development, i.e., reviewing existing methodology, devising a plan to determine water-movement rates in mountain soils, and selecting radioactive tracers for water and testing their compatibility with typical soils; (2) test methodology, i.e., measuring water-movement rates under controlled conditions and testing methods for saturated vs. less-than-saturated conditions; (3) developing a computerized accounting and storage of data and preliminary data reduction; and (4) preparing a manual for laboratory and field water-tracer studies.

Techniques for Use of Radioisotopes in Tracing Reservoir Leakage. AT(49-11)-2800. Bureau of Reclamation. This project is concerned with developing new methods of investigating reservoir sites where the problem of leakage might exist. Definite advantages can be attained by the use of radioisotopes for detecting and tracing such leakage. Where contamination by groundwater or ancient water has occurred, the radioisotope method is often the only practical means of deriving volumetric and directional flow information. For tracing leakage in complex and faulted or

cavernous strata, such as may occur in foundations and abutments, the radioisotope method can be adapted to continuous reading by detecting the actual movement of the tracer past the point of measurement.

OCEANOGRAPHY

Application of Nuclear Techniques to Marine Minerals Technology. AT(49-11)-2775. U. S. Department of the Interior, Bureau of Mines. The contractor's marine minerals research program includes a study to determine the feasibility of nuclear and radioisotope techniques for both the delineation and characterization of marine mineral deposits and the control of mining operations. Thus far the effort has been concentrated on in situ activation analysis techniques. Elemental analyses were performed on four types of sea-floor minerals of potentially commercial value: phosphorite, manganese nodules, sand (spiked with gold), and red clay. Elemental concentrations were determined by characteristic radiations emitted after irradiation by neutrons of different energies and at different flux levels.

The project was intended to serve as a start for a long-range program designed to produce, by about 1970, a Mineral Detection Probe. This instrument package will be built to probe, from sea floor to bedrock, the vertical dimension of an unconsolidated mineral deposit and provide at least semiqualitative and, hopefully, quantitative analyses. The system will not require the drilling of a hole prior to use; therefore the speed of evaluating a submerged beach or stream placer mineral deposit will be greatly increased and the total exploration and evaluation cost considerably decreased. For this reason the probe is thought to have eventual commercial possibilities.

Addition of Sensors to Gamma Radiation Spectrometer. AT(49-11)-2839. U. S. Naval Ordnance Laboratory. The project is intended to provide oceanographic data by relating radioactivity measurements to ocean conditions. Sensors were added to the Gamma Radiation Spectrometer to measure depth as a function of pressure, temperature at varying depth, and to provide information for identifying the water mass. Radioactive sources were used in the depth and temperature sensors. Thus unused channels in the multichannel analyzer could be used to provide a record of pressure and temperature as a part of the digital readout system. All data come through a common readout system.

During the past year the additional sensors were fabricated and tested using ^{133}Ba as the radioactive source. Tests were conducted both at the Naval Ordnance Laboratory and at sea.

Currently the testing program is continuing with studies using ^{204}Tl in the temperature sensor and ^{57}Co in the pressure sensor. Ultimately the detector fitted with the two sensors will provide data in actual oceanographic use.

Nuclear Oceanographic Techniques. AT(49-11)-3000. Naval Oceanographic Office, Research and Development Department. A jointly sponsored program exists for the evaluation of proposals and for experimentation with various isotopic techniques for the measurement of oceanographic parameters. Previous unofficial cooperation led to the Deep Water Isotope Current Analyzer and the Nuclear Sediment Density Meter. The Naval Oceanographic Office will:

1. Conduct studies and experiments on the applications of isotopic techniques to the measurement of oceanographic parameters. The studies and experiments will be conducted both in the laboratory under controlled conditions and at sea under operating conditions. In addition, some isotopic techniques and systems from other DID contractors will be introduced into this joint program.
2. Conduct studies and experiments on the Nuclear Sediment Density Meter in order that it can be modified to best meet Navy requirements.
3. Reanalyze and evaluate the Deep Water Isotope Current Analyzer in order to determine its full potential in naval oceanographic applications. Comparisons with conventional current sensors will be made.
4. Assist DID in evaluating new oceanographic proposals.
5. Exchange information with DID in fostering new techniques.
6. Make recommendations for Navy utilization of isotopic techniques found to advance the state of the art.
7. Assist DID in maintaining liaison with other branches of the Navy for planning conferences, etc., both classified and unclassified.

Radioisotopic Tracer Study To Investigate the Mechanics of Littoral Transport Around Point Conception, California (RIST). AT(49-11)-2988. U. S. Army, Coastal Engineering Research Center. A study to develop techniques for measuring sediment movement in the littoral zone is specifically designated for the Point Conception area of the Pacific Coast. The basic program includes development of a radiation monitoring system capable of deep-water monitoring of sand tagged with radioisotopes; analysis of data obtained from measurement of the tagged sand that has been dispersed by normal waves, tides, and alongshore currents; development of methods for tagging sand without altering its hydraulic properties; and a hazards evaluation of the entire transport measuring system.

The system is adaptable to sediment transport studies in both fresh and salt water to depths of 175 fathoms. The radioisotope used is ^{133}Xe, which gives no significant radiation hazard and no alteration of the hydraulic properties of sand indigenous to the test area. Data collection from scintillation-type detectors is obtained from punched tape and digital readout from a 400-channel analyzer. Data may be

simultaneously coordinated with depth, salinity, and temperature during the monitoring operation.

Several federal and state agencies, U. S. Army, Corps of Engineers; U. S. Navy, Pacific Missile Range; U. S. Air Force, First Strategic Aerospace Division, SAC; National Aeronautics and Space Administration; U. S. Atomic Energy Commission, Oak Ridge National Laboratory; and State of California, Department of Water Resources, are cooperating in the program.

POLLUTION

Use of Radioisotopes in Process Control. AT(29-2)-1355. Colorado School of Mines Research Foundation, Inc. Counting systems using radioisotopes and suitable for process control were developed and evaluated. Liquid scintillators were found best for most applications. Demonstration units were placed in both a large potash mill where control was achieved by counting the ^{40}K isotope and a chromium plating operation for counting ^{51}Cr in a process stream. Calculations also show that isotopes can be used to control processes for molybdenum extraction, zinc refining, and rare-earth extraction. Systems were not actually installed in these operations for reasons unrelated to the capabilities of the counters or the isotopes.

Studies on Application of Radioisotope Techniques in Stream-Pollution Problems in the Pulp and Paper Industry. AT(45-1)-1951. College of Engineering, Research Division, Washington State University. Previous research has shown that paper pulp can be successfully tagged with iridium; iridium is ideal since it is not normally present in pulp and paper mill systems, and it can be activated to produce ^{192}Ir as a tracer with an appropriate half-life for the application. Current effort is directed toward research on those problems which the Northwest Pulp and Paper Association deems most urgent to reduce the amount of waste material which enters the public waterways. The work will include all steps from laboratory through actual testing in the plant systems. In addition, attempts will be made to trace additives, such as filler clays, TiO_2, alum, and sizing agents, using the technique of activation analysis.

Studies of the Measurement of Sulfur Dioxide in Stack Gases. AT(30-1)-3881. Nuclear Science & Engineering Corporation.* The objective of this research project is to study the feasibility of a radioisotope system to measure the SO_2 content of gas effluents from power plants. The study, which will compare the technical and economic parameters of approximately six prospective methods, takes into consideration the requirements to provide data descriptive of the total effluent gas stream and the sulfur dioxide content thereof, and to provide continuous measurements with the accuracies and sensitivities required by industry. It is anticipated that the initial feasibility study will permit selection of one or two prospective systems for detailed technical and economic evaluation. This total system evaluation is expected to result in a conceptual design of a system that is competitive with existing or prospective alternatives, is compatible with the adverse environment of industrial stacks, and has capability for continuous operation without jeopardizing plant operating efficiency.

Studies of the Measurement of Sulfur Dioxide in Stack Gases. AT(30-1)-3882. Industrial Nucleonics Corporation.* Studies are being made to determine the feasibility of and to evaluate various radioisotope methods for measuring sulfur dioxide in stack gases. The studies will include direct measurements, such as the selective absorption method of low-energy X or gamma rays and X-ray fluorescence techniques, and indirect measurements, such as radio-release methods, Kryptonate techniques, and elemental exchange methods, coupled with low-energy X- or gamma-ray techniques.

The significant progress in the use of low-energy X or gamma sources of radioisotopes for analytical purposes has made it possible to apply these techniques to quantitative analyses of various elements in an adverse environment. The capabilities of Kryptonate methods and radio-release techniques for sulfur dioxide detection have already been proved. Analyses will be made to define the degree of interference introduced by other constituents of stack gases or by the need for a continuous supply of fresh radioisotopes. The high concentration of sulfur dioxide in stack gases may make it possible to detect high-atomic-number elements released through an elemental exchange process by means of an X-ray fluorescence or selective-absorption technique.

The studies will include survey and analysis of various nuclear techniques, and selection, evaluation, conceptual design, and cost estimation of the recommended system. The accuracy of measurement is to be within ±15% of the expected sulfur dioxide concentration range of 500 to 4000 ppm. The studies will be made with a 6-man-month effort ending Dec. 31, 1967.

MISCELLANEOUS

Compilation of Beta-Excited X-Ray Source Spectra. AT(11-1)-1239 [original is AT(11-1)-1593], Modifications Nos. 1 and 3. Edsel B. Ford Institute for Medical Research. This work was instituted to determine comparative spectrums for three main types of beta-excited X-ray sources—apposition, mixture, and compound. This was done in sets, each consisting of a beta source and a series of target elements, activity being held constant for comparison purposes.

*A third contract to study the feasibility of developing a nuclear gage to assist in combating this important natural problem has recently been assigned to Texas Nuclear Corporation. It is described on page 83 of this issue.

Other pertinent data will be presented with the compilation of each spectrum. Detection equipment was standardized with a 3- by 3-in. NaI crystal and a low-energy proportional counter. Spectrums were determined by standard methods. Calibrated sources were used to establish peak energies, and emphasis was placed on the lower energy characteristic peak determinations with the proportional counter. These spectrums will be assembled with the related data. Part I of the project, consisting of about 1600 spectrums, has been completed, and about 600 of these are covered in Part I of the report (TID-22361) and include 31 target elements of ^{32}P, ^{45}Ca, and ^{147}Pm. Parts II and III of the report will follow.

A Study of Fluorescent Excitation Using Isotopic X-Ray Emission. AT(40-1)-3224. Texas Nuclear Corporation. This research will develop X-ray measuring techniques in the long-wavelength region below an energy of 1 kev using thin-window, external, low-pressure chambers and other techniques and devices. The use of Ge(Li) and other solid-state detectors for X-ray detection and measurement will be investigated in the energy region below 1 kev. The magnitude of beta excitation in various materials will be investigated and compared with photon excitation. Techniques will be developed for using the additional sensitivity provided by isotopic beta excitation, especially in the very low energy region. Ten portable isotopic X-ray fluorescence survey meters, based on the techniques developed in this work, will be constructed. These instruments will be evaluated with the cooperation of industrial firms in actual industrial applications and uses.

The Study of Nuclear Resonance Fluorescence with Compton-Scattered Gamma Rays from a Gamma Monochromator. AT(40-1)-3410. Texas A & M Research Foundation. A gamma monochromator has been built for the study of nuclear resonance fluorescence. The ^{60}Co source, the aluminum scatterer, and the target to be studied all lie on a reference circle. Because of this geometrical arrangement, all gamma rays from the source that are scattered by the aluminum and passed through the target, have scattered through the same angle. Therefore all the gamma rays passing through the target have the same energy, and the desired monoenergetic gamma-ray beam has been obtained. The energy of the beam striking the target is easily varied by moving the target along the reference circle. An energy variation from about 350 kev to about 1.1 Mev can be obtained (shielding requirements limit the scattering angle to values greater than about 20°). Some applications for such a variable energy gamma-ray source are: calibration of gamma-ray detectors, solid-state research, nuclear physics research, and analysis of materials. The nondestructive identification of materials can also be carried out with the instrument.

Demonstration of Capabilities of Beta-Excited X-Ray Sources. AT(11-1)-578, Proj. No. 30. IIT Research Institute. The contractor will procure the components to fabricate and assemble one each ^{147}Pm and ^{90}Sr source, demonstrate to interested potential users the capability of these sources, and provide DID headquarters with a semiannual report of demonstrations and a list of individuals and organizations to whom demonstrated. The work will be done as outlined in proposal 66-514A dated Feb. 21, 1966.

Feasibility of Isotopic X-Ray Sources for Portable Stress-Analysis Equipment. AT(11-1)-578, Proj. No. 28. IIT Research Institute. The feasibility of using an available ^{55}Fe X-ray source with Polaroid film for measuring stress analysis under field conditions is being investigated. Further improvement in speed and resolution can be achieved for general diffraction work, but the diffraction angles are poorly situated for stress work. With pulse-counting equipment, however, diffraction lines from iron have been determined to within 0.1 to 0.2° in a few minutes' time with elementary equipment, which is capable of considerable improvement.

A Study of the Generation and Practical Uses of Monoenergetic X-Rays Using Alpha-Emitting Radioisotopes. AT(30-1)-3491. Panametrics, Inc. The objective of the work is to study the production rates of monoenergetic X rays by alpha radiation on target material and to compare the observed rate with the theoretical. Theoretical and experimental studies include measurements of sensitivity and specificity of detection, integrity of sources, matrix effects, interfering radiations, and errors caused by environment.

This study is directed toward a solution to the problem of determining the contaminants in engine oil. Additionally, the study should include the feasibility of using the X-ray sources as a method for other analytical problems.

Application of Alpha Spectrometry to Measurement of Natural Radioactivity. AT(30-1)-3335. Lamont Geological Observatory. The Atomic Energy Commission has loaned to the Lamont Geochemical Laboratory a TMC model 402 multichannel analyzer with readout and spectrum stripping modules. This unit, which is operated continuously, provides two-thirds of the analyzer time of the laboratory.

A number of tasks have been set forth in the contract: (1) techniques for high-resolution spectrometry of uranium and thorium; (2) solid-source radium spectrometry; (3) isotope dilution measurement of ^{210}Po; and (4) studies of the geochemistry of uranium, thorium, and radium isotopes in natural materials.

Continuing Research on Surface-Contoured, Semiconductor Radiation Detectors. AT(30-1)-3246. General Electric Company, Space Sciences Laboratory. Avalanching, or internal amplification, in silicon junc-

tion detectors is made possible by surface shaping, and this signal enhancement can be used to detect soft X rays at room temperature. The contoured-detector tunnel-diode detection circuit is insensitive to leakage currents generated in any way, even those greater than 100 μa. The contoured detector has three parts: (1) a dead region, 1 or 2 μ thick, at the entrance surface, where ionized carrier will invariably recombine before being usefully collected; (2) a "drift" region, where carriers generated by ionizing events are collected by the small field of 50 volts/cm due to the gradient of diffused acceptors; and (3) the multiplying, or junction, space-charge region, where carrier multiplication occurs due to the charge generated here or "drifted in" from the drift collector region. Carrier recombination in the drift region along with the loss in the dead surface region constitutes the effective window of the detector. Dimpling or etching a depression into the p^+ region to provide the proper drift-region thickness for particle detection reduces the noise level to that of the external amplifier, even to within a few volts of avalanche breakdown, and makes the particle-drift region thinner without adverse injection effects. Nearly the full 5.45-Mev alpha charge can be collected from the drift region without application of external bias. The 1.49-kev aluminum X ray, the 5.41-kev ^{55}Fe X ray, and the 9.6-kev ^{55}Fe photon have been detected by absorption in a contoured-detector structure. The large signal output of the contoured detector could be used with an external-charge-sensitive amplifier in constructing a workable medical probe aimed at detecting 5- to 15-kev X rays.

Development of Neutron Transmutation Doping of Semiconductors. AT(30-1)-3124. Fundamental Methods Associates, Inc. The objective of this program is to advance the technology of single-component fabrication to microcircuit fabrication. The basic technology of transmutation doping has been mastered, and the electronic components (transistors, diodes, resistors, and capacitors) used in microcircuits have been made by transmutation doping, all with acceptable electronic properties. Several simple microcircuits have been made. The transmutation doping process has not yet been used to fabricate microcircuits with the functional capabilities now current in the semiconductor industry, and this must be demonstrated. These microcircuits must be exhibited in packaged and operational form and fabricated in sufficient number to provide data on performance, cost, and reliability so that equipment users will feel sufficient confidence in the process to consider using it.

Research and Development on Single-Crystal High-Resistivity Cadmium Telluride for Use as a Gamma-Ray Spectrometer. AT(04-3)-549. Hughes Research Laboratories. Cadmium telluride (CdTe), with a wide band-gap energy of 1.4 Mev, a high average atomic number of ~50, and a potentially long minority carrier lifetime, is a prime candidate for use in gamma spectrometry devices requiring distinguishable photopeaks and good resolution at room temperature. Starting material of high resistivity (~10^8 ohm-cm), high carrier mobility, and low trapping rate is required for use in crystal or bulk counters. Trapping due to impurities and structural defects is the dominant factor in limiting the performance of high resistivity p-type cadmium telluride. A modified Bridgman technique, growth from the vapor phase, and a Czochralski pulling technique were used in preparing CdTe single crystals. The Bridgman technique gave CdTe crystals of both p- and n-type conductivities with resistivities of n-type material from a low of 2.5 ohm-cm to a high of 10^6 ohm-cm, and with resistivities of p-type material in the range 10^7 to 10^9 ohm-cm. Hole mobility was as high as 750 cm^2/(volt)(sec), with an average of 100 to 250 cm^2/(volt)(sec). The better crystals averaged 50 to 100 dislocations/cm^2, as revealed by an etch-pit technique. Magnesium chloride was found suitable for compensating p-type CdTe, while boric acid was suitable for doping n-type crystals. The results of depletion layer capacitance and current–voltage measurements in surface barrier devices (Al, Cu) showed the presence of nonuniformities, which acted as carrier-trapping centers. The minority carrier lifetime was much shorter than the recombination lifetime computed from theory.

Development of Methods of Industrial-Control Measurements of Low-Level Contamination of Materials. AT(30-1)-3540. Department of Physics, Franklin and Marshall College. The objective of this study is to develop practical methods of industrial-control measurements of low-level contamination of materials, i.e., counting methods that are sensitive enough to distinguish between contaminated and uncontaminated materials and are simple and economical enough for use in an average industrial laboratory, with emphasis on use of commercial equipment that is suitable or readily adaptable. Major effort will be on the laboratory proof of the measurement techniques and preparation of manuscripts containing specifications and instructions in sufficient detail to permit an industrial technician to set up the apparatus and perform the measurements in his own laboratory. Based on recent theoretical and research results, the technology of determining lead with very low levels of radioactive contamination will be developed.

Construction of a Low-Background Beta-Detector System for Solid Sample Assay. AT(49-11)-2955. Marine Radioactivity Laboratory. This program is intended to provide for the construction, calibration, and standardization of a low-background beta-detector system for solid sample assays of certain radionuclides that are found in small amounts in seawater. The system will be used for the determination of radionuclides in seawater samples from Arctic Ocean

stations collected by the Marine Radioactivity Laboratory. In addition, it will be used for analysis of samples taken from exploratory cruises to various parts of the earth.

Kryptonate Applications Investigation—Hydrogen Detection System. AT(30-1)-3735. Panametrics, Inc. The main effort of this program is concerned with the design and construction of a hydrogen detector based on the well-established Kryptonate technique. An all-effluent counting-type system will be used together with homogeneously Kryptonated platinum dioxide. Basically, this system will consist of a packed cell of Kryptonated platinum dioxide coated with a molecular barrier-type material such as aluminum oxide. Ambient air will be drawn through the cell by means of a constant-volume pump. The Kryptonate sensor cell will be operated at a constant temperature of 100°C to eliminate temperature fluctuations. Hydrogen existing in the ambient air will react with the platinum dioxide, releasing gaseous ^{85}Kr. The radioactive gas is then pumped through a counting chamber and the activity detected with a radiation detector. To meet all the requirements of this program most satisfactorily, a solid-state detector rather than a thin-window beta detector will be used. Such a system would provide a minimum effluent counting rate of 2×10^3 counts/min for 0.25% hydrogen or, since the response is linear with hydrogen concentration, 8×10^4 counts/min for 10.0% hydrogen.

Mössbauer Spectrometry for Analysis of Iron Compounds. AT(40-1)-3673. Nuclear Science & Engineering Corporation. This project is designed to explore and evaluate the potential of Mössbauer spectroscopy, using the scatter technique as an analytical tool for examination of metallurgical structures occurring in iron and steel products. The program will include the following phases:

Phase 1.

a. Determine the feasibility of constructing a Mössbauer spectrometer employing the scatter (backscatter) technique for laboratory use in determining each of the following structural characteristics of iron and steel products:

(1) The volume fraction of each phase in a multiphase mixture
(2) The kinetics of strain aging in iron products
(3) Corapositional and structural patterns for identification of metals

b. If a device for use in laboratories is found to be feasible, determine the feasibility of constructing such a spectrometer for plant and field applications.

Phase 2.

a. Design and construct a laboratory-type spectrometer suitable for demonstrating the areas of application(s) identified in *Phase 1.a.* that appear feasible for Mössbauer analysis.

b. With the laboratory-type spectrometer constructed in *Phase 2.a.*, experimentally evaluate and compare the sensitivity and utility of the Mössbauer (scatter) measurements with that of other measurement techniques now in use, such as X-ray diffraction.

Investigation of Neutron Activation for Process Control. AT(40-1)-2980. Texas Nuclear Corporation. This research is an extension of an earlier contract under which a neutron generator and electronic data-analysis equipment were developed to investigate the use of neutron activation techniques for continuous product analysis. The objective is the development of a system capable of monitoring the flow of solid or slurry products to provide the continuous analysis required for typical quality-control programs. Experiments will be conducted using simulated product flow conditions to determine the accuracy of continuous measurement of ash and heat content of coal, copper in ores, nitrogen and phosphorus in fertilizers, iron-to-silicon ratio in iron ore, pharmaceutical constituents, and other applications to be determined. All practical means of signal enhancement will be investigated including coincidence, anticoincidence, and time-of-flight detection of prompt and delayed radiations resulting from both pulsed and continuous beam neutron activation. The investigation will include economic evaluation of the system and comparison with competitive monitoring techniques. In addition, for each process application deemed feasible, a practical evaluation of potential health hazards associated with the process, including any possibly induced radioactivity in the product, will be made. This health-hazard evaluation will include data generally required as backup information for product licensing.

Liquid Loop for Activation Analysis. AT(29-2)-2115. Kaman Nuclear Division, Kaman Aircraft Corp. Liquid in a loop system is irradiated with neutrons, and the induced radioactivity is counted on a multichannel analyzer as a measure of the salt or metal content in the solution. The system should be flexible, simple, easy to install, and compatible with existing industrial process equipment. The induced activities should be short-lived. In aqueous systems considerable amounts of $^{16}N(T_{\frac{1}{2}} = 7.35$ sec) are formed by the $^{16}O(n,p)^{16}N$ reaction, but this activity can be allowed to decay in most cases before counting the sample.

Irradiation of simulated seawater at a neutron flux of 1.5×10^{10} n/sec for a period of 5 min showed distinct gamma peaks at 1.63 Mev due to $^{20}F[(T_{\frac{1}{2}} = 11$ sec) from the $^{23}Na(n,\alpha)^{20}F$ reaction] and at 3.1 Mev due to $^{37}S[(T_{\frac{1}{2}} = 5.0$ min) from the $^{37}Cl(n,p)^{37}S$ reaction]. Either of these peaks could be used for the continuous analysis of seawater for salinity down to levels of 0.02%.

Work will be done to determine the sodium-to-phosphorus ratio by neutron activation of detergents, especially the new nonfoaming types. A relative precision of 0.1 to 0.5% is required. Some of the metallic impurities in boiler water and lubricating oils will be measured by neutron activation to see whether the detection sensitivity is adequate.

Computer-Coupled Activation Analysis. AT(40-1)-2671. Texas A & M Research Foundation. Neutron activation analysis has been used to solve a number of problems of scientific and public interest. The multicomponent gamma-ray spectrum of an activated sample is unscrambled by spectrum stripping and the use of a least-squares fit of the data. A Ge(Li) solid-state detector can be used to obtain improved resolution of gamma peaks where necessary, and coincidence-counting techniques are useful when interference from other activities is encountered. Pesticide residues containing bromine, arsenic, or mercury, in the parts per million range, were analyzed in various samples of hay, foliage, fruit, and vegetables submitted by the U. S. Department of Agriculture. A method of removing sodium by precipitation with HCl–butanol was developed to improve the accuracy of analyzing biological samples for trace metals. The liver of a child who died of selenium poisoning was found to contain about 0.5 μg of selenium per gram of liver. Ore samples containing vanadium, titanium, rare earths, or aluminum were easily analyzed by neutron activation analysis.

The light elements, especially O, N, C, B, and F, can be selectively analyzed by bombarding the sample with high-energy ^{3}He ions on an 88-in. variable-energy cyclotron. Particle energies of 6 to 55 Mev for protons, 12 to 65 Mev for deuterons, 18 to 120 Mev for ^{3}He ions, and 25 to 130 Mev for ^{4}He particles can be obtained. Charged-particle activation analysis will be difficult to evaluate because of the rapid loss in energy as the charged particle penetrates the sample.

Basic Development and Application of Bubble Chambers for Low-Level Counting. AT(30-1)-3506. William H. Johnston Laboratories, Inc. Current work is aimed at establishing the feasibility of the bubble chamber for low-level measurement of ^{14}C and ^{3}H. Background will be reduced by the use of Pyrex glass instead of quartz and an anticoincidence guard counter. The entire chamber will be designed to increase the volume and to permit the incorporation of components for rapid recompression and sensitive detection. The effects of impurities and different fillings on bubble-chamber performance will be studied.

Development of Krypton-85 as a Universal Tracer. AT(30-1)-2757. Panametrics, Inc. The purpose of this contract is to develop methods for incorporation of ^{85}Kr into solids that will retain the ^{85}Kr and to observe the properties of such preparations to determine conditions of release and release rates in order that the controlled release can be used in making physical measurements such as for temperature and pressure. The contract period, December 1964–November 1966, has two purposes: (1) to maintain a facility for preparation of Kryptonated materials and to provide consulting services to government agencies and industry and (2) to investigate applications of Kryptonation technology previously developed specifically to determine (*a*) utility in wear studies, (*b*) utility in phosphors, (*c*) surface temperatures of components subject to supersonic velocities, and (*d*) detection of hazardous gases and degradation of thermal-control coatings.

A Study To Improve Radioisotope-Excited Light-Source Techniques. AT(30-1)-3816. Rensselaer Polytechnic Institute. In this work the use of radionuclides to produce luminescence is being investigated. Special cells containing multiple windows will be constructed to contain ^{85}Kr and tritium mixed with other gases. The emission spectrum from one window will be observed from the region of 1050 to 15,000 A, and the other windows will be covered with phosphorescent material. The intensity of the visible light emitted will be measured with a photomultiplier and the luminescent light-emission spectrum determined with a spectrograph. Initial experiments will examine the spectrum for the emission of the resonance lines of Ar, Kr, Xe, H, and admixtures of He–N, Ar–N, etc. The phosphorescent materials to be examined include the NBS standards: zinc sulfide (with activators Ag, Cu, Au, and coactivators Al, Ga, In, and Cl), CdS, ZnSe, CdSe, and other inorganics, and pure and doped anthracene, naphthalene, phenanthrene, and other organic phosphors. Some cell modifications to use liquid scintillators will be made.

Until the receipt of radioactive gases and the construction of the specialized vessels, the initial research will make use of the cells and the ^{210}Po alpha source presently available, using pure noble gases and admixtures of the noble gases. The light output from the phosphorescent materials will also be examined as a function of temperature. Breakdown of fluorescing material due to ionizing radiation from tritium-containing compounds or ^{85}Kr gas has been the chief problem experienced to date.

Radiochemistry Techniques in Evaluation and Improvement of Industry Practices for Removal of Pesticide Residues from Food. AT(04-3)-536. National Canners Association. The aim of this study is to investigate by use of tagged radioactive pesticides the removal of pesticides from food and to improve methods of pesticide removal. Radioactive pesticides such as DDT-^{14}C, DDT-^{3}H, malathion-^{14}C, and diazinon have been used to study the fate of pesticides in foods, to devise better pesticide extraction schemes, to determine pesticide distribution in the plant, and to evaluate the effectiveness of detergent and water washes. The use of radioactive DDT-^{14}C gave results accurate to the 0.1 ppm pesticide level. The pickup of DDT-^{14}C from the soil decreased with carrot age: from 0.99 ppm DDT for 64-day carrots to 0.24 ppm for 124-day carrots. A total of 68% of initial DDT was found, by scintillation counting, in the hair roots, tap root, and carrot skin, i.e., portions that are discarded during

food preparation. Autoradiograms of carrots grown in DDT-^{14}C-treated soil showed migration of the DDT from the roots to the foliage, but the activity pickup (0.4 to 1.0 ppm DDT) was well below the established tolerance of 7 ppm for DDT in carrots. Extraction of soil with dichloromethane gave a complete recovery of 10 ppm of DDT-^{14}C from the soil. Autoradiographs showed homogeneous distribution of DDT-^{14}C in the soil. Work on the extraction and clean up of pesticide residues is being extended.

In past work, fruits (peaches, pears, and strawberries) and vegetables (corn, broccoli, spinach, and tomatoes) were sprayed with ^{134}Cs solution. Most of the applied radioactivity was located on the outer parts of the plant with small amounts in the flesh. Immediate washing prevented migration of radioactivity from the skin to flesh. The distribution of ^{134}Cs in various parts of the plant was shown for plants grown on contaminated soil. Spinach roots contained a larger amount of radioactivity than the stems. Broccoli leaves showed twice as much activity in the leaves as was present in the heads. Corn skins showed more activity than the milk. Zirconium phosphate ion exchanger was effective in reducing the ^{134}Cs content in tomato juice to a value below background.

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Radiation Processing Applications Development

Electron and X- and Gamma-Ray Research for Radiation Processing. AT(49-2)-1165. U. S. Department of Commerce. The main purpose of this project is development and field testing of a chemical dosimetry system suitable for industrial applications in the radiation processing of foodstuffs, plastics, medical supplies, etc. Such a system would have the following desirable characteristics: (1) a linear response between optical transmittance density and radiation dose in the range of 10^3 to 10^7 rads; (2) a low dependence on temperature and on energy and intensity of the radiation; (3) a high reproducibility and stability factor and good spatial resolution; and (4) simplicity and convenience in preparation and use, and usefulness over a wide range of industrial applications.

Results of tests using radiation sources similar to those which would be used in industry (electron beams and ^{137}Cs and ^{60}Co gamma-ray sources) show that certain colorless dye precursors, which may be used in a variety of forms such as thin films, gel blocks, and liquid cells, take on deep colors upon irradiation. Dose response upon irradiation is determined by relating the radiation dose to the corresponding increase in optical density measured at a given wavelength of light.

In addition to the dosimetry studies, studies of techniques for energy measurement of monoenergetic electron beams are under way. Supplementary work includes coordination with other National Bureau of Standards (NBS) projects (for example, the Berger–Seltzer computations of photon and electron transport) and currently informing selected representatives of manufacturers concerned with radiation-processing equipment of related NBS activities.

Proposal for Survey of Potential MISER Applications. AT(30-1)-3736. Atlantic Research Corporation. A four-phase program was conducted on a survey of potential applications of microbiological inhibition systems employing radiation (MISER). Phase I was a state-of-the-art evaluation and involved a literature compilation. Phase II involved a broad evaluation questionnaire that requested information regarding microbiological control presently being performed and also requested an indication of willingness to participate further in programs of radiation testing. The mailing list included investigators currently active in the field, industrial groups, government, and universities and associated hospitals. Phase III was an in-depth survey to select promising MISER applications, and Phase IV was a detailed evaluation of specific systems and applications to determine technical and economic feasibility. Patented areas of MISER applications were industrial cutting fluids, jet fuels, and cosmetics. USAEC Report* NYO-3736-2 gives details of the completed program. As a result of this study, a continuation of the effort on cutting fluids has been initiated with NUMEC.

Vectored Research on Promising Radiation Systems. AT(30-1)-3655. William H. Johnston Laboratories, Inc. Irradiation of a system with gamma radiation or high-energy electrons can produce radicals that will combine with each other or react with other components of the system to produce novel compounds. A Van de Graaff accelerator is available to give 400-kev electrons. Irradiations are also performed with an 8-kilocurie ^{60}Co source, or a 600-curie ^{90}Sr source. After irradiation the samples are analyzed by gas chromatography or on a mass spectrometer to determine G yields or to identify the reaction products over a wide range of reaction conditions. Tetramethylmethylenediamine (TMMD) was produced at G yields of 383 to 425 by irradiating dimethylamine (DMA) containing 4.7 to 7.4% O_2 with a dose of 5.2 Mrads of ^{60}Co gamma rays at a temperature of 220°C and a pressure of 3.0 to 3.6 atm. Dimethylethylsilane was obtained at a G yield of 15 without nitric oxide being present and at a G yield of 8 in the presence of nitric oxide (a free-radical scavenger) by irradiating a mixture of dimethylsilane and ethylene. On irradiating a mixture of methysilane and ethylene, the G yield for the formation of methylethylsilane was 33 with nitric oxide present and 39 without nitric oxide. At a higher temperature of 200°C and a pressure of 10 atm, the G value for methylethylsilane was 292 in the run with 4.6% nitric oxide, 239 in the run with 3.6% NH_3, and 403 with neither nitric oxide nor NH_3 present. Methylethylsulfide was obtained at a G yield of 800 to 1800 (yield being proportional to the pressure) by irradiating a mixture of methylmercaptan and ethylene with 400-kev electrons from the Van de Graaff at a temperature of 110°C, a total pressure of 2.5 to 9.8 atm, and a mole ratio of 3 to 1 excess ethylene. Methylpropyl sulfide was produced at a G yield of 870 to 1950 on substituting propylene for ethylene.

Research and Development Studies into the Basic Technology of Radiation-Induced Ionic Polymerization Including Small-Scale Pilot-Plant Systems. AT(40-1)-3667. North Carolina State University. Research will be continued on new radiation-induced ionic polymerization and copolymerization systems, and a small semicontinuous pilot plant will be constructed to study selected systems under more realistic conditions. The source to be used is the 27,500-curie ^{60}Co source at North Carolina State University. Among the variables which need investigation with regard to problems which may arise during scale-up

*Available from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151.

are heat transfer, residence-time distribution in real systems, and the effect of agitation, i.e., in continuous-flow systems. More controversial is the establishment of extreme dryness in the reaction system under small-scale semicontinuous-flow conditions. A number of methods are available and will be studied from the point of view of both G values and reproducibility.

Styrene and/or α-methyl styrene will be used initially for these studies; however, monomers such as trioxane, vinyl chloride, 4-methyl pentene, and others will be included in later studies. The studies will be performed in a reactor system that will permit continuous sampling of the material in the storage tank and the effluent from the reactor zone so as to enable careful evaluation of reaction kinetics.

Development Studies for a Radioactive Pebble-Bed Chemical Reactor System. Phase II. AT(30-1)-3864. Research Department, Western New York Nuclear Research Center, Inc. The objective of this research program is to investigate the feasibility of radiation-induced side-chain chlorination of toluene and then whether or not the chlorination process can be adapted to a radioactive pebble-containing chemical reactor system. A pebble-bed reactor system is one in which the chemical reactants would be circulated through a reaction vessel containing a very large number of small radioactive beads or pebbles exposing the reactants more effectively to the radiation field to initiate a long-chain (high-G-value) chemical reaction. The use of ionizing radiation to replace the usual ultraviolet-light initiation would eliminate problems associated with highly light-absorptive reactants and would permit increases in the effective volume of the reaction vessel.

The present research will study the effect, on the benzotrichloride yield (G value), of (1) temperature, (2) flow rate of both gas and liquid, (3) surface-to-volume ratio, and (4) benzal dichloride. The radiation will be 0.7-Mev electrons produced by a Van de Graaff accelerator. Dose rate will be varied from 10^4 to 10^6 rads/hr, and temperature will be varied from 0° to 100°C. The reaction would be adaptable to ionizing radiation processing only if the G value is high (indicating a chain-reaction free-radical process). Since minimal pebble-bed attrition is required for the pebble-bed concept to be commercially attractive, tests will be made, concurrent with the chlorination study, in a fluidized system using reaction vessels constructed according to several designs developed in the Phase I study of this program [AT(30-1)-3718]. Inactive test pebbles will be fabricated to contain a "stable" tracer such as yttrium, europium, or dysprosium, and tests of fluidization velocity, pebble size, pebble mass, and solution volume will be carried out in the reaction vessels. After each test, neutron activation analysis, preceded by suitable preactivation concentration steps, will be carried out to determine pebble attrition.

The results of these parallel studies of the chemical reaction and the reactor system will determine the future of the concept.

Development of a Radioactive Pebble-Bed Chemical Reactor System. Phase I. AT(30-1)-3718. Western New York Nuclear Research Center, Inc. In a previous theoretical feasibility study, the use of radioisotopes in the photochemical production of benzotrichloride from toluene and chlorine was shown to be competitive with, or even more economical than, the presently used light sources. The variables in this reaction and the mechanics of a suitable pebble bed on a small, laboratory scale will be studied now. Variables to be considered are temperature, dose rate, chlorine and toluene flow rates, the concentrations of benzotrichloride and side products, pebble size and materials, and pebble attrition. Van de Graaff electrons will be used to simulate the effect of radioactive pebbles.

The goal of this effort is to find a set of operating conditions and to design a pebble-bed reactor that will give the most economical production of benzotrichloride on a commercial scale. This will permit direct comparison between the potential economics of the radiation-initiated and the photochemically initiated processes.

Use of Nuclear Radiation in the Preparation of Optical Laser Materials. AT(30-1)-3330. Radioptics, Inc. This contract extends the research performed under an earlier contract to enhance the performance of laser materials through the use of nuclear radiation. In the previous work, rods of ruby, neodymium-doped barium silicate glass, and neodymium-doped soda-lime glass were exposed to 2.4×10^7 rads of ^{60}Co radiation. The rods were shown to have little or no laser output immediately after irradiation. However, after controlled thermal and optical bleaching of the radiation-induced color centers, the ruby rod and the barium silicate glass rod surpassed their original unirradiated performance. The soda-lime silicate rod did not reach original performance even after annealing at 300°C for 2 hr, but further annealing is expected to improve its performance. The improved performance in ruby and in barium silicate glass is quite stable under operating conditions. Color centers that remain after 300°C annealing are deep-seated, and their lifetimes at normal operating temperatures will be long.

Current research will include an evaluation of the radiation process with respect to composition of the material, radiation dosage and dose rate, irradiation temperature, various bleaching and annealing conditions and cycles, and the temperature of laser operation after irradiation.

Other material compositions to be investigated are lanthanum and borate-base glasses, selected rare-

earth dopants in glass hosts, and neodymium in single crystal calcium tungstate. The effects of ultraviolet and infrared bleaching will be determined as well as the effect of elevated annealing temperatures.

Radiation Synthesis of Artificial Kidney Membranes. AT(30-1)-3329. Columbia University, Department of Chemical Engineering. The objective of this project was the synthesis of artificial kidney membranes by the radiation cross-linking of polyvinyl alcohol (PVA) and polyvinyl pyrrolidone (PVP). The two polymers were cross-linked very efficiently by the use of small amounts of allyl methacrylate in conjunction with radiation. The PVP cross-linked membranes were not particularly useful as artificial kidney membranes because they were physically too weak to even allow the testing of permeability properties. The PVA membranes were tested in the dialyzer and possessed good permeability characteristics. However, their strength is low, and efforts to improve this property have been unsuccessful. Radiation alone or in combination with monomers, however, offers two satisfactory ways of cross-linking polymers. Applications of radiation to polymers of greater inherent physical strength may prove more fruitful for the synthesis of artificial kidney membranes. A final report is being printed.

Industrial Evaluation of Radiation-Treated Wood–Plastic Materials. AT(40-1)-3415. Lockheed-Georgia Co. This project involved a cooperative government–industry program for evaluation of radiation processed wood–plastic composites (WPC) in a variety of end-product applications. Seventy-eight companies representing a broad spectrum of products were selected by the Atomic Energy Commission for participation in the program.

Wood supplied by each participant was converted to WPC at the Lockheed-Georgia Nuclear Laboratory and returned to the participant for product evaluation. Consulting services were provided to all participants throughout the program. Over 8000 lb of WPC involving 41 different woods was supplied. The evaluations, which were received from the 68 participants who supplied wood for processing, included data on such characteristics of the WPC as stability and ability to absorb moisture, to be machined, and to be glued, nailed, etc.

Development of a Radiation Process for Wood–Plastic Combinations. AT(40-1)-2945. Modification No. 10. West Virginia University. Radiation polymerization of vinyl monomers for use in WPC will involve studies of (1) copolymer systems, e.g., styrene/acrylonitrile and styrene/vinylidine chloride; in addition, new copolymer systems will be sought and kinetics data will be analyzed; (2) polymer systems containing plasticizers to determine whether or not these systems have mechanical properties measurably different from those of unplasticized WPC; (3) inert additives to reduce raw-material cost and liberated heat of polymerization; (4) cross-linking agents for dose reduction; (5) kinetics of styrene/acrylonitrile polymerization at 10,000 rads/hr; and (6) polymerization at varying dose rates extended to include styrene/acrylonitrile and ethyl acrylate/acrylonitrile. Engineering support will involve continued studies of impregnation variables, adiabatic polymerization, heat transfer, and uniform and nonuniform (surface) monomer loadings. Evaluation of WPC will involve mechanical properties, fire and chemical resistance, microscopic studies of polymer distribution, moisture diffusion, decay and termite resistance, and dye fading.

An Investigation of Certain Physical and Mechanical Properties of Wood–Plastic Materials. AT(40-1)-2513, Task 20. Research Triangle Institute. This work assesses the nature and extent of changes in selected physical and mechanical properties of irradiation-produced WPC as a function of the monomer loading. In a subcontract program with the North Carolina State School of Forestry and the West Virginia University Engineering Experimental Station, the Research Triangle Institute is responsible for liaison, statistical design of the study, and analysis of the results. Physical and mechanical tests, e.g., static bend, hardness, and toughness, will be made on samples of various combinations of wood species, monomers, and monomer loading. The tests will give information on the following sources of variation: differences between two trees of the same species, between vertical positions in the same tree, and between homogeneous groups (replication) within logs; tree-to-monomer loading and log-height-to-monomer loading interactions. Over 5000 tests will be required for the program. Test data will be tabulated for transfer to IBM punch cards for data analysis using the RTI Bunker-Ramo computer at Duke University.

The Radiation-Induced Polymerization and Copolymerization of Unusual Monomers. AT(40-1)-2513, Task 13. Research Triangle Institute. Research work on the radiation-induced polymerization of unusual monomer and comonomer systems is terminating. Detailed studies have included (1) the attempted copolymerization of a number of materials, e.g., trithiane and β-trithiobenzaldehyde, in the solid state; (2) the copolymerization of sulfur dioxide and various vinyl monomers, e.g., terpolymerization of isoprene–sulfur dioxide complex and the butadiene–sulfur dioxide complexes with a number of monomers that do not themselves polymerize with sulfur dioxide; and (3) the copolymerization of formaldehyde in the liquid state with vinyl and other comonomers, e.g., vinyl ethyl ether and butadiene.

Phase diagrams have been made for various mixtures of monomers to find the optimum conditions for solid-state copolymerization. The study of sulfur dioxide terpolymers has been extended to long-chain

methacrylates and acrylates such as 2-ethyl-hexyl acrylate. Thermal gravimetric analysis has been used to screen all aldehyde-vinyl copolymer samples prepared to date, and other monomers — *p*-methyl styrene, *p*-nitro styrene, etc. — as comonomers for formaldehyde have been investigated.

Radiation-Induced Emulsion Polymerization. AT(40-1)-2513, Task 17. Research Triangle Institute. This program has illustrated the advantages of radiation as a polymerization initiator for emulsions, i.e., latex systems. Emulsion polymerization is an excellent technique for the synthesis of high polymers. A unique advantage of radiation-induced polymerization is that no chemical initiators are required — initiation may be controlled by dose rate alone, independently of temperature. Thus disadvantages of chemical initiators for emulsion polymerization are avoided. The use of radiation to produce high-molecular-weight latexes of polyvinyl acetate at 0°C and other temperatures has been investigated, and the latex properties, rates of polymerization, and molecular weights have been studied. The extent of the "aftereffect" during the emulsion polymerization of vinyl acetate, styrene, and other monomers has been measured at various temperatures. Butadiene and isoprene have been polymerized in emulsions using radiation initiation, and several solvent and transition metal compounds have been added and the effect on structure determined.

Copolymers of Tetrafluoroethylene for Superior Dielectric Materials. AT(30-1)-2982. Harry Diamond Laboratories, Department of the Army; RAI Research Corp. Tetrafluoroethylene was copolymerized with ethylene, propylene, 4-methylpentene-1, or styrene by irradiation to obtain products with improved dielectric properties. Under rigorously anhydrous conditions, a total dose of 2×10^6 rads has given copolymer yields equivalent to those obtained with a dose of 10×10^6 rads under nonanhydrous conditions. About 7 g of ethylene-tetrafluoroethylene (E-TFE) copolymer (23.4% conversion) was obtained at a dose of 2×10^6 rads. Tensile strength and Young's modulus of the E-TFE copolymer at ambient temperature were 5560 psi and 4.47×10^5 psi, respectively, compared to 2000 to 4500 psi and 0.58×10^5 psi for Teflon. Tetrafluoroethylene was also copolymerized with propylene monomer under anhydrous conditions, as shown by the similarity of infrared spectrums of the ethylene and propylene copolymers, and data were accumulated to determine the molecular weight of the copolymer by this method.

The Effects of Ionizing Radiation on Colloids. AT(29-2)-1653. Unidynamics, Phoenix. A preliminary study of the effects of ionizing radiation on colloidal systems of special interest to various industries was made. Goals were the disclosure of radiation effects that might have industrial applications and selection of areas best suited for more study.

Three colloidal systems and a class of chemicals used industrially to prepare emulsions were evaluated. The principal findings were

1. Radiation induces significant changes in the rheological properties of aqueous sols of a number of polymers. Some of these (notably polyethylene oxides) exhibit changes at radiation doses which are biologically significant.

2. Irradiation significantly affects the stability of aqueous suspensions of bentonite clays. These effects are dependent upon dose rate, total dose, and solution environment in which the irradiation is performed.

3. Significant radiation-induced changes occur in the chemical and physical properties of certain nonionic surfactants.

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Radiation Preservation of Foods

Petition Preparation and Submission. AT(30-1)-3135. Hazleton Laboratories, Inc. Petitions will be prepared and submitted to the U. S. Food and Drug Administration (FDA) establishing the safety of certain irradiated food products. The number of petitions to be prepared and the food products to be covered will be in accordance with a program prescribed by the Commission, and a draft of each petition will be

submitted to the Commission for review and approval prior to submission to the FDA. Efforts on collection and review of pertinent data will be coordinated with such primary sources as Massachusetts Institute of Technology; U. S. Army Quartermaster Corps, Natick Laboratories; Bureau of Fisheries of the Department of Commerce; various Commission contractors; various U. S. Army contractors; and foreign sources. A chart will be prepared ranking food items for petition preparation, and tentative target dates for submission of petitions will be determined, based on research status.

Commercial Prospects for Selected Irradiated Foods. AT(49-11)-2797. U. S. Department of Commerce. Under direction of the U. S. Department of Commerce, the Battelle Memorial Institute has completed a study to analyze and evaluate the commercial prospects of certain irradiated foods by an intensive study of economic, marketing, and production factors. Courses of action that could expedite commercialization are recommended. A report for food-industry management was prepared. Foods included are poultry, marine products, bananas, sterilized pork and beef, and dehydrated vegetables.

Cost-Benefit Study of Selected Products in the Atomic Energy Commission's Low-Dose Food Irradiation Program. AT(30-1)-3666. Daniel Yankelovich, Inc. This firm undertook a study of six items in the AEC's low-dose food-irradiation program to determine benefits that could be expected from continued research support and to evaluate consumer attitudes toward irradiated foods. On the basis of 310 interviews with industry personnel and 627 interviews with consumers, and as a result of a study of government research project reports, suggestions were made as to areas for continued research and appropriate funding levels were recommended. It was suggested that the AEC should give firm support to research on irradiation of New England finfish and Hawaiian papayas, marginal support to Hawaiian mangoes, and no further support to Dungeness crab, Alaskan king crab, and bananas. As a result of the consumer survey, it was concluded that over 80% of the consumers will buy irradiated food if it is approved by the FDA and marketed under well-known brand names.

Effects of Low-Level Irradiation upon the Preservation of Food Products. AT(40-1)-3097. Agricultural Experiment Station, University of Florida. The effects of gamma radiation on citrus fruits, mangoes, peaches, strawberries, and tomatoes will be determined. Heat-treatments in combination with irradiation will be investigated to determine if this procedure will permit use of lower radiation doses for spoilage control. Organoleptic quality will be evaluated, and physical and biochemical changes will be measured. Since previous research has shown that the response of fruit species to gamma irradiation varies according to variety, maturity, and climatic conditions prior to and at the time of harvest, the dosage that the fruit can tolerate as related to these variables must be determined. Postirradiation variables of temperature and packaging and the effect on fruits will be studied, as will changes in respiration, ethylene production, pigment development, sugars, organic acids, and flavor components. Texture modifications as related to pectin degradation will be determined. Some shipping tests of citrus fruit will be conducted.

Simulated Commercial Shipping Studies. AT(49-11)-2954. U. S. Department of the Interior, Bureau of Commercial Fisheries. Studies of simulated commercial shipping conditions will be made in support of petitions. "Good," "current," and "abuse" conditions, based on present and projected industrial practices will be duplicated as closely as possible and will take into account initial handling, processing, irradiation, shipping, and storage variables.

Cost-Benefit Analysis of Selected Pasteurized Meats. AT(30-1)-3831. Daniel Yankelovich, Inc. The basic objective of this contract is to evaluate and estimate both tangible (e.g., benefits from improved economic efficiency) and intangible (e.g., improved public health) benefits likely to accrue as a result of commercialized radiation pasteurization of fresh meats. The evaluation is to take into account (1) the value added by the process, (2) the relative costs of radiation and other techniques for achieving the same purposes, (3) the relative market acceptability of these several techniques, (4) the expenditure of government funds required to achieve commercialization, and (5) the costs to be incurred by private industry in establishing and operating the process.

Radiation Pasteurization of Fresh Meats. AT(11-1)-1689. Food Science Department, Michigan State University. The perishability of fresh meats (<4-day display life) requires meats to be cut and packaged in the retail store. The centralization of this processing would offer the possibility of significant cost reductions. A doubling of the life of fresh meats by improved handling would not be sufficient to permit centralized cutting. Hence some means in addition to improved handling are needed. The research planned will involve the combination of irradiation with other agents to attain a fully satisfactory extension of the life of fresh meats. The studies will be concerned with (1) the use of ionizing radiation to control spoilage due to microbial action and (2) the use of appropriate agents to control color deterioration and fluid exudation.

Irradiation Treatment of Grain and Grain Products. AT(49-11)-2676. U. S. Department of Agriculture, Stored Products Research Laboratory. Research on the radiation treatment of grain and grain products will be coordinated. The effects of radiation on insects and mites infesting grain as well as on the stored product

itself will be determined. These studies should result in establishment of definitive dosages and should elucidate such problems as radioresistance. The bulk grain and packaged commodity irradiator located at the Stored Products Insect Research and Development Laboratory at Savannah, Ga., will be used for these studies.

Commercial Storageability of Irradiated Potatoes (original title, Engineering and Economic Feasibility). AT(11-1)-1539. Pillsbury Co. The Pillsbury Co. has conducted an experimental program with two varieties of potatoes to compare the effects of irradiation and chemical treatment on a group of potatoes as compared to an untreated or control group. Test and control groups, consisting of about 36 thousand lb of potatoes each, were stored for various periods of time, then withdrawn from storage, analyzed, and tested for processing characteristics. Potato processing companies other than Pillsbury may participate in this phase at the discretion of Pillsbury. A final report is being prepared at this time.

Shelf-Life Extension and Quarantine Control of Tropical Fruits by Irradiation. AT(04-3)-235. University of Hawaii, College of Tropical Agriculture, Hawaii Agricultural Experiment Station. For disinfestation of many tropical fruits and vegetables by ionizing radiation, dosages may be as low as 15 krads, and 30 krads has been provisionally accepted as adequate for a wide spectrum of products. Many advantages over the fumigation methods now used are inherent in the irradiation process; furthermore, fumigation methods are totally ineffective on some products, e.g., mango and avocado. Radiation control of fruit flies in papaya, avocado, and other fruits and of seed weevils in mango is of particular commercial interest and potentiality in Hawaii. Specific objectives include determination of (1) organoleptic, shipping, and storage qualities of irradiation-disinfested papaya and the effects on these qualities of the interaction of heat-disinfestation with radiation-disinfestation treatments; (2) nutritive and selected biochemical properties, after suitable storage, of radiation-disinfested mango; (3) effects of product irradiation on the virulence of postharvest decay fungi, principally on mango and papaya; (4) nutritive and organoleptic qualities and disinfestation effectiveness of low-dose gamma treatment of pineapple, lychee, avocado, pepper, tropical fruit juices, and selected tropical fruits; (5) market acceptability and development and pre- and postharvest handling of irradiated papaya.

Radiation Technology in Conjunction with Postharvest Procedures as a Means of Extending the Shelf Life of Fruits and Vegetables. AT(11-1)-34. University of California, Department of Pomology. Major emphasis will be placed on the combined effects of heat and irradiation on market quality of nectarines and peaches. Studies will be made to define precisely the optimum levels of heat and irradiation for rot control, with minimum injury to the fruit, by comparing the effects of heat, irradiation, and combination treatments with unirradiated, unheated fruits with respect to organoleptic attributes, flavor, texture, rate of ripening, ascorbic acid content, and susceptibility to mechanical injury. Simulated and actual test shipments will be performed on treated fruits that show promise for rot control without major adverse effect on fruit quality.

Packaging studies will be made in an attempt to minimize mechanical injury of irradiated cantaloupes, nectarines, peaches, and tomatoes during marketing. Actual transcontinental test shipments will be attempted (time permitting) with irradiated nectarines, peaches, strawberries, cherries, figs, bushberries (blackberries, dewberries, and raspberries), and oranges processed by the Mobile Gamma Irradiator. Studies will be made on quantitatively inoculated cantaloupes to determine the radiation sensitivity of the major melon pathogens. Mushroom studies will be continued to evaluate precisely the effects of gamma irradiation on storage and shelf life.

Investigations relating to fungicidal treatments will concentrate on the combined effects of heat and irradiation. If disease conditions are favorable, an attempt will be made to evaluate for peaches the benefits of the combined treatments following natural orchard infections. Hopefully, heating and irradiation dosage recommendations will be forthcoming. The possibilities of combining heat with radiation in the treatment of strawberries will be further explored. The response of sweet cherries and selected vegetables to combined treatment will be investigated.

Irradiation of Fruits and Vegetables. AT(11-1)-1592. Department of Food Science, Michigan State University. As a continuation of research already in progress, a number of fruits and vegetables, including strawberries, sweet cherries, peaches, and tomatoes, will be irradiated to reduce microbial contamination and extend marketing life. Attempts will be made to overcome the radiation-induced softening of fruits by dipping them in tissue-firming solutions. The delay of maturation of certain vegetables after harvesting will be further explored. The combination of irradiation and heating will be tried as a preservation process for certain fruit juices.

Effect of Irradiation on the Microbial Flora Surviving Irradiation-Pasteurization of Seafoods. AT(04-3)-502. Oregon State University. The metabolic and spoilage characteristics of radiation-surviving microorganisms will be investigated by standard microbiological procedures, by inoculation into sterile fish muscle, and by manometric techniques. The major radiation survivors belong to genus *Achromobacter*, and investigation of this group will be emphasized. However, *Lactobacillus* and "coryneforms" are also included in this study.

Survival characteristics of *Salmonella* species in inoculated seafoods are to be investigated. The mutagenic effect of radiation to this group and other radiation-surviving microorganisms will be determined.

Chemical food preservatives that have been shown to increase the lethal effect of radiation will be studied. Work with sodium benzoate, potassium sorbate, tylosin lactate, and EDTA is also included in this study.

Radiopasteurization of Fishery Products and MPDI Operation and Developmental Investigations. AT(49-11)-1889; AT(49-7)-2443. U. S. Department of the Interior, Bureau of Commercial Fisheries, Gloucester. These contracts entail the investigation of radiopasteurization of fishery products of commercial significance and the establishment of handling and processing methods necessary for commercial application of radiation in the seafood industry. The work is divided into two parts: (1) laboratory research and (2) developmental studies involving the Marine Products Development Irradiator (MPDI).

The major objectives of the research are to (1) establish the maximum shelf life of radiopasteurized and nonirradiated haddock and codfish fillets (based solely on the odor of the raw samples); (2) determine the optimum dosage for radiopasteurizing oysters, shrimp, and blue-crab meat; (3) continue the study of the feasibility of using radiopasteurization to extend the shelf life of "slacked-out" products (products stored and handled frozen until they reach the retail outlet, whereupon they are thawed and held refrigerated); (4) complete the study of the relationship of preirradiation quality on postirradiation quality and shelf life of clam meats and cod fillets, and to survey the preirradiation quality of ocean perch relative to its postirradiation shelf life; (5) continue to relate organoleptic changes in radiopasteurized, radiosterilized, and nonirradiated fish with changes that occur in the composition of the volatile chemical components in the product; and (6) monitor bacteriological quality of test samples to observe the relationships between quality and total plate counts in both irradiated and nonirradiated fish.

Commercial Irradiation of Shellfish with a Portable Shipboard Irradiator. AT(10-1)-3413. Department of Food Science and Technology, Louisiana State University. The portable shipboard irradiator will be used for making commercial feasibility studies at a dockside shrimp unloading site on the Gulf coast. Shrimp that have been washed, headed, and iced-stored will be obtained from several trawlers to uncover problems created by variations in ship construction and handling by different crews. At the dock the shrimp will be packaged and irradiated at 0.22 ± 0.05 Mrad and iced-stored for appraising shelf-life extension. Products showing evidence of decomposition will not be irradiated. The samples will be analyzed chemically for indole and ammonia and microbiologically for total counts and several pathogens. Organoleptic evaluations will include general appearance, odor, texture, sweetness, flavor, and black spot.

Results will be reviewed for possible application to commercial practices and for solving unforeseen problems that could serve to aid in the preparation of a petition to the FDA for approving low-dose radiation preservation of shrimp.

Application of Radiation Pasteurization Processes to Pacific Crab and Flounder. AT(49-11)-2058. U. S. Department of the Interior, Bureau of Commercial Fisheries. The objective of this work is to develop a practical procedure for the radiation pasteurization of Pacific crab meat, flounder fillets, and related products. The specific tasks are (1) to determine the optimum pre- and postirradiation conditions (sample treatments, radiation dosage, storage-life tests, etc.) directed toward defining commercial process parameters; (2) to develop methods for maintaining quality control and for establishing the safety of the radiation-pasteurized seafoods by using sensory, microbiological, chemical, and physical tests; (3) to determine the maximum safe storage time ("X" value) on the basis of unanimous rejection of irradiated fish fillets by an untrained panel. This time will be related to the toxin production in fish fillets first inoculated with spores of *Clostridium botulinum* type E and then irradiated and stored at various temperatures for comparable times to establish the safety of the irradiation process; and (4) to determine the commercial feasibility of the radiation-pasteurization process for industry by shipping commercial quantities of irradiated fillets through regular commercial channels.

The Effect of Gamma Rays on Cod Fillets Inoculated with Type E, Cl. Botulinum. AT(30-1)-3343. Department of Food Technology, Massachusetts Institute of Technology. The D values (a measure of the efficiency of spore reduction) will be measured in a mixed culture in cod at 10^2, 10^4, and 10^6 spore levels to determine whether minced cod flesh reacts in the same way as minced haddock flesh. Upon completion of this and similar studies, if it has been demonstrated that a relationship does exist, an extended inoculated pack study will be run. In the planned study, cod fillets will be inoculated with 1×10^3 spores per gram of type E, *C. botulinum*, packed aerobically and anaerobically, irradiated at levels of 150, 250, and 350 krads and stored at 33, 35, 40, and 45° F for approximately 1, 2, and 3 times the irradiated storage life at these temperatures, and finally tested for outgrowth and toxin production.

It is expected that this investigation will provide information concerning the suitability of low-fat fish as a growth substrate for type E, *C. botulinum*. This investigation would also determine whether low-fat fish, when irradiation pasteurized, present a particular problem from the standpoint of type E botulism.

Since anaerobic packs of fish inoculated with type E, *C. botulinum*, irradiation pasteurized, and stored have not been examined for outgrowth and toxin production, this investigation would provide information concerning any hazards that might be associated with the vacuum packaging of irradiation-pasteurized marine products.

Radiation Pasteurization of Shrimp and Oysters. AT(40-1)-2951. Department of Food Science and Technology, Louisiana State University. Radiation pasteurization of shrimp and oysters has proved to be a highly successful procedure for increasing their iced-storage life. Physical, chemical, microbiological, and organoleptic studies have been completed and show that this process is safe and that, when subjected to consumer appraisal, irradiated shellfish are preferred over the nonirradiated products.

The scope of proposed research will include investigations into the value of reduced temperature during radiation (cryogenics), the required condition of shellfish prior to irradiation, mechanisms by which texture seems to be improved during irradiation, the development of a simple procedure for determining the extent of radiation, the effect of radiation on traces of insecticidal residues that may be found in shellfish, and the isolation and identification of compounds and their changes during radiation. Microbiological techniques, chemical analyses, gas chromatography, infrared spectrum analysis, organoleptic tests, and nutritional evaluations will be included in making these studies.

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Soviet-Bloc Countries Now Covered by New AEC General License for Exports

Albania, Bulgaria, Czechoslovakia, Estonia, Latvia, Lithuania, Outer Mongolia, and the Soviet Union are covered by the new USAEC general license that permits shipment of radioisotopes as labeled compounds without specific application to the USAEC. Under Part 36 of the USAEC regulations, "Export and Import of Byproduct Material," radioisotopes having an atomic number from 3 to 83, inclusive, may be exported as labeled compounds in quantities not to exceed 1 curie per shipment. Tritium in quantities not to exceed 10 curies per shipment with a specific activity of not more than 10 curies/g may be exported as labeled compounds to Poland and Romania in addition to the above countries. Exports of radioisotopes of other elements are already covered by Section 36.21 of Part 36 of the USAEC regulations.

This licensing action will reduce the administrative task of the government and of industrial concerns that sell these isotopes. This action is consistent with U. S. Government policy to reduce export controls on East/West trade in nonstrategic items.

New Hanford Contracts

Atlantic Richfield Co. of Philadelphia has been selected to operate the reactor fuel processing plant at Hanford, near Richland, Wash., which will recover uranium and plutonium from spent fuel. This is a renegotiation of the Isochem contract, which included building a $\$8.5 \times 10^6$ fission-product recovery plant with private funds. In the Atlantic Richfield proposal there was no specific commitment to build a fission-product recovery plant.

NUMEC Testing Cardiac Pacemaker

The first model of a cardiac pacemaker, an electrical pulse generator being developed for the medical

treatment of heart block, is being tested by Nuclear Materials and Equipment Corp., a USAEC contractor. The current model uses conventional electricity, but the eventual product is expected to be powered with ^{238}Pu. Operation will probably be at a very low power level — about 160 μw.

Navigational Satellite Marks Sixth Anniversary

The first nuclear power generator in space, designed for 5 years' operation, marked its sixth anniversary on June 29. The grapefruit-sized 5-lb 3-watt nuclear generator, developed for the USAEC by Martin Co., Baltimore, was installed to supplement solar power on the Navy's 175-lb drum-shaped navigational satellite. It contains ^{238}Pu.

New Documentary Motion Picture Features BNL

"Brookhaven Spectrum" is a new 25-min color motion picture that surveys, in nontechnical terms, some of the nuclear research and engineering at the USAEC's Brookhaven National Laboratory. Available for free loan or for purchase, the documentary film is of interest to the general public, including high school students and service clubs. Among the research projects reviewed are: preservation of perishable foods with high-intensity radiation, autoradiography of rare oil paintings by neutron activation, aging-mechanism studies made with radiation, and a newly developed treatment for leukemia patients by extracorporeal irradiation of the blood.

Contract for Designing Meat-Irradiation Pilot Plant

The USAEC has selected Irradiated Foods, Inc. (IRRADCO) of Allentown, Pa. to design, build, and operate, with Government cooperation, a pilot plant for preserving meat by irradiation. IRRADCO is co-owned by Isotopes, Inc., of Westwood, N. J.; Allen Products Co., Inc., of Allentown, Pa.; Martin Marietta Corp., Baltimore; and Uniroyal, Inc., of New York. The plant is expected to be operational in about 2 years. It will use 400 thousand curies of ^{60}Co from Neutron Products, Inc., of Washington, D. C., and its initial annual capacity will be 3 million lb of meat, including ham, pork, chicken, and beef.

Hawaii Development Irradiator Dedicated

The Hawaii Development Irradiator installation, a pilot plant for processing tropical fruits with ^{60}Co gamma radiation, was dedicated in Honolulu on July 3. Processing of fruits for preservation, spoilage reduction, and disinfestation to overcome quarantine barriers and facilitate marketing on the mainland will be studied. Initial emphasis will be on payayas and mangoes.

The Hawaii Development Irradiator will also share in the Defense Department's wheat gamma-irradiation program, with Lockheed-Georgia the other participant. These two groups will process 130 thousand lb of wheat flour for disinfestation purposes.

Texas Nuclear Awarded DID Contract To Study Measurement of Sulfur Dioxide in Stack Gases

Texas Nuclear Corporation has been awarded AEC research contract No. AT(30-1)-3880, the objective of which is to survey possible nuclear methods for determining SO_2 in stack gases and to conduct feasibility studies of methods that show promise. Results of the feasibility studies will be used in the selection of one or more systems for further evaluation. The evaluation studies will include consideration of the following factors: (1) compatibility with environment, (2) interference with plant operation, and (3) competitiveness with other types of systems. The contractor will prepare a final report recommending a program for the development of the selected system or systems. (Two earlier contracts with Nuclear Science and Engineering Corporation and Industrial Nucleonics Corporation are described on page 69 of this issue.)

Conferences

IAEA and Michigan State University Present Food-Irradiation Course

Representatives of 20 countries have completed an 8-week course in food irradiation at the Michigan State University, which cosponsored the course with the International Atomic Energy Agency (IAEA). The International Training Course on Food-Irradiation Technology and Techniques, which ran from June 18 to August 11, provided intensive training for scientists who already have backgrounds in irradiation, food technology, agriculture, forestry, fisheries, or nonmedical aspects of human nutrition.

The course, directed by Professor Walter M. Urbain of Michigan State University's Department of Food Science, covered the following topics: fundamentals of radiation; radiation chemistry; effects of radiation on living organisms; dosimetry; radiological safety; irradiation; packaging, storage, wholesomeness, and government regulation of irradiated foods; commercial outlook and considerations for irradiated food; interest and activity concerning food irradiation in various nations; current and future problem areas; and literature sources.

Countries represented at the course were Argentina, Australia, Brazil, Bulgaria, Chile, Republic of China, Czechoslovakia, Greece, India, Iran, Israel, Italy, Peru, Philippines, Poland, Romania, Spain, Thailand, Turkey, and Venezuela. Two of the attendees, L. Turjanski of Argentina and E. Eisenberg of Israel, are already quite active in food irradiation. Nearly every researcher already well known for contributions to the subject participated in the lecture program.

IAEA Symposium on Radiosterilization of Medical Products Features Code of Standard Practice

During the week of June 5–9, 1967, nearly 200 scientists from 26 countries met in Budapest for the IAEA-sponsored Symposium on Radiosterilization of Medical Products. Recent research work assessing the usefulness of radiation over other sterilization methods was reported, and the first code of practice for radiation sterilization of medical products was presented and discussed.

The code was worked on for 2 years by various IAEA panels and working groups of experts; it should greatly assist countries not yet having legislation on the use of radiation-sterilized products. The first general principle of the code is the need for wide safety margins; an important point is the inclusion of measures to minimize contamination at all stages of manufacture. Radiation doses should be graded according to material. Microbiological standard preparations would come from IAEA, and checks on effectiveness would be made by using deliberately contaminated and infected articles. One of the chief advantages of radiation sterilization is that it is a "cold" treatment; this has allowed the use of new materials such as plastics in manufacture and packaging. Kallings (Sweden) pointed out that the timing of the code is appropriate because of the current state of development and the spread of radiation sterilization.

Surgery to replace parts of the human body, such as bones and heart valves, has had much success using radiation to kill harmful germs in the implanted material. Antibiotics, injections, human blood plasma, thyroid substances, catgut, and eye solutions, among other substances, have been investigated.

Studies reported from Hungary and the United Kingdom showed that infective germs do not appear to like plastic materials. There have been good results in heart operations using plastic tubes and artificial valves in patients. Bone banks in Poland and Austria use radiation to sterilize specimens; Ostrowski (Poland) said that over 6 thousand specimens were irradiated in Warsaw and all were sterile when tested later for bacteria.

Availability of Isotopes and Services

ORNL Stable Isotope Inventory Augmented

Recent additions to the ORNL enriched-stable-isotope inventory include the following:

Isotope	Purity, %	Amount available, g
^{28}Si	99.9	15.1
^{66}Zn	98.5	51.6
^{90}Zr	97.7	267
^{106}Cd	88.4	1.5
^{116}Cd	94.3	5.0
^{116}Cd	97.2	2.0
^{152}Gd	51.0	0.2
^{196}Hg	47.6	0.4
^{208}Pb	99.5	27.6

General

Newfield Irradiator To Continue Operation Under AECL Sponsorship

The food-irradiation plant of Newfield Products Ltd., at St. Hilaire, Quebec, will be operated by Atomic Energy of Canada Ltd. (AECL) as the center of an expanded radiopasteurization program. The two foods already cleared in Canada—potatoes and onions—will be processed for sale to the public, and AECL will do research on mushrooms, strawberries, sugar beets, apples, and animal feed. In addition, AECL plans to investigate poultry irradiation in conjunction with the USAEC. When sufficient evidence is gathered, AECL intends to submit several petitions to the appropriate government authorities for clearances of other irradiated foods.

USSR Clears Irradiated Foods

The use of radiation to inhibit sprouting of potatoes and to reduce spoilage of fruits, vegetables, and dried fruits has been approved by the USSR. The dose is to be 100 to 300 krads. In addition, radiation pasteurization of meat, with a maximum dose of 500 krads, has been approved. The low-dose irradiation of meat is not yet considered economic in the United States, and little work has been done in this field.

Army and Air Force Recommend Irradiated Bacon for Use

As a result of evaluations made during December 1966, both the Air Force and the Army maintain that canned, sliced, irradiated bacon is superior to the currently issued canned, prefried bacon. The Air Force recommends that the irradiated bacon

be authorized for use in lieu of the present bacon in B rations. The Army recommends that the radiation program be continued and expanded to include other meat rations, such as hot dogs.

The Air Force issued 5448 lb of the irradiated bacon to 15 bases in the Atlanta Depot area for evaluation by mess personnel. Detailed veterinary questionnaires were returned from 13 bases, food-service questionnaires from 14 bases, and preference surveys from 12 bases. There was considerable evidence that the bacon did not comply with Air Force product characteristic requirements; slices were too wide or too narrow, and length of slice varied too much. Some bases reported that grilling gave a poor result. For the first test, 2196 airmen were served irradiated bacon; they rated it as 5.86 on the usual 9-point hedonic scale. For the second test, 1966 airmen were served irradiated bacon, and they rated it as 6.50; however, the second time they were not told it was irradiated bacon. The rating for bacon overall was 7.62 in a food-preference survey taken in 1963.

The Army served about 13,500 lb of irradiated bacon at two installations—Fort Benning and Fort Gordon—during December 1966. Almost 85% of the dining halls ovenbaked their bacon; this is perhaps the reason only 12% reported difficulty in preparing it. The 2187 soldiers at Fort Benning rated the unidentified bacon at 6.46 on the 9-point hedonic scale; on the second test, when they were told the bacon was irradiated, 2605 soldiers rated the bacon only slightly lower, at 6.38. The difference was larger at Fort Gordon; 1000 soldiers rated the bacon as 6.33 the first test but only 5.96 the second time—when they were told it was irradiated. The Army concluded that serving irradiated bacon is no different from serving unirradiated bacon; the test results were called "quite favorable."

Promethium-147 Heat Source Available

Atomics International, of Canoga Park, Calif., is offering a new heat source, called ThermoPac. It is fueled with ^{147}Pm and produces 0.5 to 5 watts(t). It has a useful life of about a year and costs from $800 to $8000.

Useful Plant Mutations

An improved peanut plant, developed with the use of ionizing radiation, has won the O. Max Gardner Award for Walton C. Gregory of North Carolina State University. The award is made for "contribution to the welfare of the human race" in increasing world food output.

ENEA Initiates Isotope Power Program

A joint program to develop isotopic power sources of 0.001 to 1 mw has been initiated by eight of the European Nuclear Energy Agency countries. The sources may be used for cardiac pacemakers, in telemetry, and in clocks.

Selected Current Literature*

New Edition of ORNL Radioisotope Catalog Now Available

Radioisotopes, Stable Isotopes, and Research Materials—a catalog of products and services—is now available from the Isotopes Development Center (IDC), Oak Ridge National Laboratory. The 61-page publication gives all necessary ordering information, including shipping data and regulations, with prices, for both radioactive and stable isotopes.

A supplement to the catalog is in preparation. This supplement will provide further nuclear data for many isotopes, a glossary, explanations, and a brief guide to the selection of an isotope for a given job. It is intended more for the inexperienced user of radioisotopes than for the experienced user, although the latter also might find it useful.

Table of Isotopes (6th Edition) Now Available

The sixth edition of the "bible" of isotopes—the first revision since 1958—has now been published by John Wiley & Sons, Inc., New York. *Table of Isotopes* is a summary of information about radioisotopes and nuclear structure; it was first published in *Reviews of Modern Physics* in 1940 by Glenn T. Seaborg and J. J. Livingood. Seaborg was a coauthor on all subsequent editions except this one, which is dedicated to him. The sixth edition is authored by C. Michael Lederer, Jack M. Hollander, and Isadore Perlman—all of the Lawrence Radiation Laboratory at Berkeley.

Whereas the first *Table* was 17 pages long and treated 330 radioisotopes and 284 stable isotopes, the latest edition is 650 pages long and treats more than 1400 radioactive and stable isotopes. The great growth in size is attributable not so much to the increase in number of known isotopes as to the increase in knowledge of their properties.

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